

# International Workshop on Stratospheric Aerosols: Measurements, Properties, and Effects

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STRATOSPHERIC AEROSOLS: MEASUREMENTS,  
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sponsored by the IAMAP Radiation  
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Ames Research Center  
March 27-30, 1990*

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**NASA**

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# **International Workshop on Stratospheric Aerosols: Measurements, Properties, and Effects**

*Edited by  
Rudolf Pueschel*

*Ames Research Center  
Moffett Field, California*

*Proceedings of a workshop  
sponsored by the IAMAP Radiation  
Commission and the NASA Upper  
Atmosphere Research Program at  
Ames Research Center  
March 27-30, 1990*



National Aeronautics and  
Space Administration

**Ames Research Center**  
Moffett Field, California 94035-1000

ORIGINAL PAGE  
BLACK AND WHITE PHOTOGRAPH



Back row, left to right: Darrell Goularte, Bernd Georgi, Jim Rosen, Joe Jordan, Stephen Randtke, Dennis Lane, Dan Murphy, Matt Hitchman, Guy Ferry, Bob Stevens.  
 Second row: Mercedes Holland, Rudi Pueschel, Gunthard Metzger, Mark Jacobson, Su Bowen, Terry Deshler.  
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 Front row: Pihuan Wang, John DeLuisi, Tony Hansen, Rich Turco, Leopoldo Stefanutti, Scott Elliott, Ken Snetsinger, Phil Hammer.

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## PREFACE

The existence of stratospheric aerosols has been linked to the Earth's climate since the beginning of systematic observations in the early 1960's. Indeed, models indicate that the injection of aerosols into the stratosphere by the El Chichon volcanic eruption in 1982 has resulted in a temperature forcing in the troposphere. A sustained stratospheric loading of this magnitude would definitely result in a deleterious climate change. The recent discovery of the conversion of inert halogen species into ozone-reactive radicals on the surfaces of polar stratospheric cloud aerosols, resulting in a net ozone depletion, further enhanced the importance of stratospheric aerosols to climate. The denitrification and dehydration of the Antarctic winter stratosphere by the sedimentation of type I and type II polar stratospheric cloud particles is probably the crucial element in allowing the springtime ozone depletion to expand to lower latitudes. Some researchers theorize that ozone depletion may occur in mid-latitudes on the surfaces of sulfuric acid aerosols after volcanic eruptions.

In light of the importance of stratospheric aerosols to climate, the Joint Working Group of the International Aerosol Climatology Project (IACP) in 1988 mandated the formation of a Subworking Group on Stratospheric Aerosols, and strongly recommended holding a Workshop to discuss relevant stratospheric aerosol measurement problems. Consequently, a workshop on "Stratospheric Aerosols: Measurements, Properties, Effects" was held at NASA Ames Research Center at Moffett Field, California, from March 27-30, 1990.

The objectives of the workshop were (1) to bring together experts from the fields of properties, effects and measurements of stratospheric aerosols, (2) to assess the present state of knowledge in these fields, and (3) to write a workshop report recommending improvements of in situ and remote sensing observational strategies for future national and international activities that deal with measurements of stratospheric aerosols.

The science issues that were addressed included (1) formation and evolution, (2) transport and fate, (3) effects on climate, (4) role in heterogeneous chemistry, including polar stratospheric clouds, and (5) validation of lidar and satellite remote sensing. Improvements of in situ measurement technologies in light of currently existing environmental issues related to stratospheric aerosols were discussed. Accordingly, among the specific aerosol characteristics that need to be measured with the best accuracy and specificity are the natural (background) and the volcanically enhanced aerosol (sulfuric acid and silicate particles), the exhaust of civil aviation, shuttle and supersonic aircraft operations (alumina, soot, ice), and polar stratospheric clouds (ice, condensed nitric and hydrochloric acids).

The first day and part of the second day of the workshop were devoted to oral presentations and discussions on the issues raised. The rest of the time was spent on drafting and discussing the workshop report. For this purpose, the workshop participants were split into several subgroups for discussions, and the compilation and recommendations of specific measurement technologies.

The three-and-a-half-day meeting was sponsored by the IAMAP Radiation Commission (IRC) and NASA's Upper Atmosphere Research Program. The meeting was hosted and organized by NASA Ames Research Center.



This volume contains the deliberations of the workshop. Thirty-three participants from universities, the private sector, research laboratories, government agencies in France, the Federal Republic of Germany, Italy, the People's Republic of China, the United Kingdom, and the United States of America participated in the meeting.

It is a pleasure indeed to thank the sponsors, the host, and all participants for their enthusiastic support and cooperation.

R. F. Pueschel

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## AGENDA

International Workshop on  
STRATOSPHERIC AEROSOLS: MEASUREMENTS, PROPERTIES, EFFECTS  
NASA Ames Research Center  
Moffett Field, CA 94035-1000  
27-30 March, 1990

### TUESDAY, 27 MARCH - BLDG. 201 - AUDITORIUM

**9:00** Introductions, Announcements

**9:15** Formation and Evolution of Stratospheric Aerosols

R. Turco: Overview

P. Hamill: Nucleation and growth of multicomponent aerosol particles

P. Hamill: Nitric acid aerosols at the tropical tropopause

J. Hallett: Laboratory and field studies of stratospheric aerosols: Phase changes under high supersaturations

Others

Discussion

**10:30** Transport and Fate of Stratospheric Aerosols

M. Kritz: Overview

M. Kritz: Physico-chemical processes controlling the spatial distribution of the Junge layer

M. Hitchman: Transport in the lower stratosphere

J. DeLuisi: A historical stratospheric aerosol data set

Others

Discussions

**11:45-13:30** Lunch - NASA Ames Research Center - Cafeteria

**13:30** Effects of Stratospheric Aerosols on Climate

J. Lenoble: Overview

F. P. J. Valero: Radiative effects of Arctic haze

A. Hansen: Measurements of aerosol black carbon ("soot") in remote regions and in the stratosphere

W. Su: The effects of sulfate and soot in smoke on atmospheric extinction

Others

Discussions

**15:00 Role of Stratospheric Aerosols in Heterogeneous Chemistry, including polar stratospheric clouds**

B. Toon: Overview

C. Wilson: Aerosol formation/properties in the polar stratosphere

L. Stefanutti: Polar stratospheric clouds during the Antarctic Winter 1989

J. Rosen: Backscatter and particle counter measurements of PSCs in the north polar vortex

T. Deshler: Measurements in polar stratospheric clouds over Antarctica in September 1989

A. Jayaraman: Study of aerosol scattering at stratospheric altitudes using rocket and balloon-borne spectrophotometer

Others

Discussions

**WEDNESDAY, 28 MARCH - BLDG. 201 - AUDITORIUM**

**08:00 Validation of Lidar and Satellite Remote Sensing**

P. McCormick: Overview  
Global data set and validation

P. Durkee: Aerosol characterization in support of the AVHRR optical depths algorithm

J. DeLuisi: (a) Status review of the world lidar network  
(b) some results of lidar comparisons with SAGEII

H. Jaeger: Lidar measurement at Garmisch, conversion model and midlatitude aerosol effects

J. Zhou: A lidar equipped with photon counting for measuring stratospheric aerosols

J. Lenoble: Comparison between balloon polarimetry and SAMII/SAGEII

R. Santer: Aerosol sensing from balloons using photopolarimetry

R. Stevens: Application of angular denuders to airborne HNO<sub>3</sub> measurements

B. Huebert: Aircraft sampling artifacts

B. Georgi: In situ particle measurements in the Arctic: Design and planning of aerosol sizing

D. Murphy: Aerosol mass spectrometer instrument development

Others

Discussion

**11:45-12:45 Lunch**

**13:00 Working group discussions and report writing:**

Bldg. 242 – Room 206 – Formation and Evolution

Bldg. 245 – Room 215 – Climate

Bldg. 245 – Room 141 – Heterogeneous Chemistry and Polar Stratospheric Clouds (PSCs)

Bldg. 245 – Room 136 – Satellite validation

**THURSDAY, 29 MARCH**

**08:30 Working group discussions and report writing:**

Bldg. 239 – Room B39 – Basement – Formation and Evolution

Bldg. 245 – Room 215 – Climate

Bldg. 245 – Room 141 – Heterogeneous Chemistry and PSCs

Bldg. 245 – Room 136 – Satellite validation

**11:45-12:45 Lunch – Officer's Club**

**13:00 NASA Ames Aircraft: HANGAR #1**

**14:00 Plenary Session: Status reports: BLDG. 201 – AUDITORIUM**

**15:00 Working group discussions and report writing**

Bldg. 239 – Room B39 – Basement – Formation and Evolution

Bldg. 245 – Room 215 – Climate

Bldg. 245 – Room 141 – Heterogeneous Chemistry and PSCS

Bldg. 245 – Room 136 – Satellite validation

**FRIDAY, 30 MARCH - BLDG. 201 - AUDITORIUM**

**09:00** Wrap-up of discussion and final editing

**12:00** Adjourn



## ACRONYMS

AAOE	Airborne Antarctic Ozone Experiment
AASE	Airborne Arctic Stratospheric Experiment
ACE	Aerosol Climatic Effects Program
ADS	annular denuder system
ALEX	Aerosol Lidar Experiment
ALEXIS	Atmospheric Lidar Experiment in Space
ALISA	(Cooperative project between France and the Soviet Union)
ALTO	Airborne Lidar for Tropospheric Ozone
ATLID	Atmospheric Lidar
AWI	Ames Wire Impactor
BALKAN	(Soviet project of airborne lidars)
CHEOPS	Chemistry of the Ozone Polar Stratosphere
Cl	chlorine
CH <sub>4</sub>	methane
ClONO <sub>2</sub>	chlorine nitrate
CN	condensation nuclei
CNC	Condensation Nucleus Counter
CNES	Centre National d'Etudes Spatiales
CNRS	Centre National de la Recherche Scientifique
CO	carbon monoxide
COS	carbonyl sulfide
D	deuterium

DIAL	Differential Absorption Lidar
DLR	Deutsche Forschungs und Versuchsanstalt für Luft und Raumfahrt
DMS	dimethyl sulfide
ESA	European Space Agency
F	fluorine
GCM	General Circulation Models
H	hydrogen
HBr	hydrobromic acid
HCl	hydrochloric acid
HF	hydrofluoric acid
HNO <sub>3</sub>	nitric acid
HOCl	hypochloric acid
H <sub>2</sub> O	water
H <sub>2</sub> SO <sub>4</sub>	sulfuric acid
IAMAP	International Association of Meteorology and Atmospheric Physics
IACP	International Aerosol Climatology Program
ICR	ice crystal replicator
IR	infrared radiation
IRC	International Radiation Commission
JSC	Johnson Space Center
LASA	Lidar Atmospheric Sounder and Altimeter
LEANDRE	(French project of airborne lidars)
LITE	Lidar In-Space Technology Experiment

MFS	Multiple Filter Sampler
N	nitrogen
NASA	National Aeronautics and Space Administration
NAT	nitric acid trihydrate
NOZE	National Ozone Expedition
N <sub>2</sub> O <sub>5</sub>	nitrogen pentoxide
PMS	Particle Measurement Systems, Inc.
PSC	Polar Stratospheric Cloud
PV	potential vorticity
SAGE	Stratospheric Aerosol and Gas Experiment
SAM	Stratospheric Aerosol Measurement Project
SME	Solar Mesosphere Explorer
SO <sub>2</sub>	sulfur dioxide

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## I. SCIENCE ISSUES

### 1.1 FORMATION, TRANSPORT, AND FATE

Panel members: Pat Hamill (Leader), John DeLuisi, Matt Hitchman, C. S. Kiang, Mark Kritz.

The panel reviewed the present state of understanding of the formation, transport and final form of the stratospheric aerosols, focusing primarily on the sulfate aerosol (i.e., the Junge layer) rather than polar stratospheric clouds (discussed in Section 1.3 of this report) or transient aerosol events such as volcanic veils.

#### 1.1.1 FORMATION

Although the origin of the aerosols is not completely known, it is clear that they are either formed in the stratosphere by some sort of in situ homogeneous nucleation mechanism, or by heterogeneous nucleation onto particulates (i.e., condensation nuclei (CN) injected into the stratosphere from the troposphere or from outer space (such as meteoritic debris, ion clusters)).

Much of the discussion centered on the recent observations by the University of Wyoming group of the appearance of high altitude, very small particles. These appear to be formed in the polar regions during the coldest part of winter and advected to lower latitudes. Since they do not seem to contain inclusions, it is believed that they are the product of homogeneous nucleation of sulfuric acid and water. The larger sulfate aerosol particles, however, usually do contain some sort of insoluble inclusion (or at least some material remnant which survives evaporation). It is not certain if this is an indication of heterogeneous nucleation processes or just the fact that these are aged aerosol particles which have undergone coagulation with available material.

The question was raised as to the origin of the sulfuric acid vapor, which gives rise to the new particle formation in the winter polar vortex. It is presently believed that the evaporation of mid-latitude aerosols at high altitudes (above about 30 km the temperature is too high to support the aerosol and the particles evaporate) will lead to a supply of gas phase sulfuric acid which will migrate to the poles. During the winter descent of air, this sulfuric acid is carried to lower levels. Since the temperature is low enough, nucleation will occur. Since this air is relatively particle free, coagulation will not reduce the number density too quickly. With the breakdown of the vortex, these particles are advected to the mid-latitudes and serve to maintain the sulfate layer by supplying new particles.

#### 1.1.2 TRANSPORT

Clearly the process envisaged above depends on transport. Unfortunately, there is much uncertainty regarding the meridional transport of the stratospheric gases and particles. It was suggested that the aerosols themselves could be used as tracers to try to determine transport. In the winter of 1982-83 the northward transport rate of El Chichon aerosols was observed by 6 lidars stationed between 19° and 55° latitude. The transport rate of the cloud maximum was estimated to be approximately  $0.3^\circ \text{ day}^{-1}$ , and at January 1, 1983, it was located near 30° N. Within the uncertainty of the measurements, the cloud maximum optical thickness appeared to remain constant with

latitude. On the other hand, the stratospheric aerosol concentration at 40° N began to rise a few weeks after the eruption. Nearly identical transport behavior was observed from sunphotometer data following the eruption of Agung.

This led to the suggestion that it might be possible to set up a lidar network consisting of perhaps 20 lidars over a region perhaps as large as the United States. If the system were operated in some sort of unmanned mode with identical lasers and data reduction schemes, one should be able to obtain an excellent picture of stratospheric motions. In particular, tropopause fold events could be studied. The fluxes of water vapor, sulfur compounds, and particulates into the stratosphere would be measured, as well as the downward flux of stratospheric aerosols (as possible cirrus nuclei). However, if one wishes to study small particle transport under background conditions, lidar measurements are susceptible to large errors during periods of low concentration. In addition, clouds could block most of the aerosol signals.

The mean circulation of the quasibiennial oscillation strongly affects the distribution of aerosols. The associated presence or absence of a subtropical barrier in potential vorticity (PV) will discourage or allow transport of aerosols out of the tropics by extratropical Rossby waves. The time evolution of volcanic veils may be used to infer the morphology of the PV barrier.

Aerosol composition is a strong function of air parcel history. Chemical General Circulation Models (GCMs) may soon be capable of representing the overlap of tracer tongues, but at present this is limited by computational capabilities. To form aerosols containing constituents with different geographical sources probably requires differential air mass advection in the vertical, coupled with small scale mixing due to gravity waves. GCMs are still a long way from being able to represent these small scale mixing processes adequately. Detailed lidar observations may allow adequate parameterizations to be created.

### 1.1.3 FATE

The last question considered was the final form of the aerosols. It is believed that the particles near the bottom of the aerosol layer are removed by being mixed out into tropospheric air. In general, the picture which emerged from the discussion was a tropical source for the reacting gases, a formation of large particles by growth onto the condensation nuclei (CN) produced in the winter polar vortex, and a slow average transport to higher latitudes. The particles are probably mainly removed from the stratosphere at the tropopause folds which are a major sink for stratospheric air. It was pointed out that subsidence of air in the winter poles may also account for a significant loss of stratospheric aerosol particles. Wintertime polar regions appear to be an efficient, probably one of the most important, sinks for stratospheric aerosols. For example, little if any of the aerosol expected to be injected into the stratosphere was observed in the tropics, following the eruption of the Redoubt volcano (60.5 N, 152.0 W) in Dec. 89/Jan. 90.

### 1.1.4 IMPORTANT SCIENTIFIC ISSUES

The following scientific issues (not necessarily in order of priority) should be addressed.

- a. A dedicated program of stratospheric dynamics to be used along with an observational program.

- b. Use of high-altitude long-lived balloons (1 month or longer) equipped with light solar powered transmitters tracked by satellite. This can be done inexpensively by using ozonesondes on special plastic balloons, which can easily reach 38 km..
- c. An example of a measurement comparison would be launching of balloons at different latitudes during the maximum wintertime transport period. Other comparisons could be run for special studies such as polar regions in winter.
- d. The SAGE II (Stratospheric Aerosol and Gas Experiment) satellite sensor and existing lidars can indicate where the aerosols are located. A U.S. lidar network would be useful but expensive. Unmanned lidars will present a difficult technical problem.
- e. The transport and modeling work will benefit stratospheric O<sub>3</sub> and other research as well as aerosols.
- f. Mountain waves and gravity waves can be used to evaluate processes occurring in the nucleation and formation of clouds in the upper troposphere and lower stratosphere.
- g. Analysis of descending air in the polar vortex to determine if it is bringing down sulfuric acid from higher altitudes.
- h. Measurements of gaseous sulfuric acid (H<sub>2</sub>SO<sub>4</sub>) in the stratosphere and sulfur dioxide (SO<sub>2</sub>), carbonyl sulfide (OCS), dimethyl sulfide (DMS) and other sulfur bearing gases in the upper troposphere. There is some question as to the validity of some of the older measurements.
- i. Modeling and theoretical studies should try to avoid using average values and use time varying parameters more appropriate to the phenomenon being studied. However, 3-D models are probably not too useful at this stage in their development because of the lack of small scale detail.
- j. Determination of whether or not the composition of the particles formed by upwelling of gases through the tropical tropopause is different from that of particles formed in the polar vortex.
- k. Determination of the CN budget.

## 1.2 CLIMATIC EFFECTS

Panel members: John Hallett (Leader), Tony Hansen, Jacqueline Lenoble, Wei Han Su.

This working group discussed the various aspects of the stratospheric influence on climate by identifying specific questions and topics that, in our opinion, warrant further investigation. This group believes that stratospheric aerosol investigations are vital in future research programs.

Aerosols affect the climate by changing the radiation budget. They affect the amount of radiation reaching the surface and the amount of radiation that escapes to space. The amount of heat retained or lost at different altitudes, longitudes, or latitudes in turn affects the atmospheric dynamics. The aerosols' direct radiation effect is due to the scattering, absorption, and emission properties of the



aerosol particles. The exact concentration, size distribution, composition, shape (physical state), and refractive index (homogeneous or inhomogeneous mixtures) of the aerosols around the globe results in a redistribution, in the atmosphere, of energy from solar and terrestrial radiation. Thus, atmospheric heating or cooling depends on the type and spatial distribution of the aerosols in the atmosphere; it also depends on the transition of aerosols to clouds. The importance of aerosols acting as cloud nuclei lies in controlling drop or crystal size distribution, concentration, and optical thickness of such clouds.

The stratosphere may exert a considerable influence on tropospheric climate through its effects on radiative transfer, through the circulation of particulate material in the upper atmosphere, and also through its transport down through troposphere folds to affect clouds in the upper troposphere. Therefore, any considerations of climate change must include considerations of this stratospheric component.

The effects of major perturbations to the stratosphere on planetary habitability have already been demonstrated by the following two examples. 1) Aerosols injected into the stratosphere by the El Chichon eruption may have resulted in a measurable temperature forcing in the troposphere; a sustained stratospheric aerosol loading of this magnitude would definitely result in a climate change, and 2) the destruction of ozone, mediated by polar stratospheric clouds (PSCs), may lead to increased UV flux at the surface. Hence, the importance of the stratosphere in surface climate is already documented.

## **1.2.1 CLIMATIC EFFECTS OF STRATOSPHERIC AEROSOLS**

### **1.2.1.1 Direct Radiative Effects of Clouds and Aerosols**

Clouds and aerosols suspended in the atmosphere interact with radiation by scattering and absorption. In this report we will concentrate on effects due to optical properties in the solar spectral region as well as the longwave infrared, where absorption usually dominates. In general, the albedo increases with increasing optical thickness. The albedo is affected by particle concentration, composition and phase, size distribution and morphology. In the solar spectral region, the optical properties of most natural aerosols are dominated by scattering, but anthropogenic activities may be releasing increasing quantities of optically-absorbing material to the atmosphere (e.g., soot).

### **1.2.1.2 Indirect Effects**

Changes in the temperature forcing of the stratosphere may result in changes of stratospheric circulation and exchange with the troposphere. In addition, changes in stratospheric temperature may affect the phase-change of certain key species (e.g., the formation of PSCs), which in turn will have an effect on climatically-important parameters.

From the above, we believe that the presence of aerosols in the stratosphere, from both natural background and natural perturbations such as volcanic eruptions and anthropogenic activities, may have an influence on climatically-important atmospheric parameters.

This implies that in order to understand the radiation balance at these levels and its influence on tropospheric climate, we need observations of the particulates at selected locations, altitudes, and times. These observations will provide the input data to model calculations of the sensitivity of climate to stratospheric aerosols.

### **1.2.2 EXISTING CAPABILITIES**

Existing capabilities consist of the following:

- a. Measurements of stratospheric aerosols by in situ techniques (i.e., balloons, aircraft) of concentration, composition, size distribution, morphology.
- b. Measurements of stratospheric aerosol optical depths from aircraft, balloons, and spacecraft.

For example, both aircraft and spacecraft measurements have been used to determine the temperature dependence of the formation of PSCs. Thus it was discovered that the frequency of PSC occurrence increases with decreasing temperatures below about 195 K.

### **1.2.3 PRESENT MEASUREMENTS**

Present measurements consist of the following:

- a. Balloon Soundings: Vertical profile of number count, size distribution (limited knowledge), backscatter, optical depth, polarization.
- b. Aircraft: Limited altitude measurement of number count, chemical composition of individual particles, size distribution, optical depth.
- c. Lidar: Vertical profiles of aerosol backscatter and depolarization.
- d. Satellites: Wide-area vertical profiles of aerosol optical extinction.

### **1.2.4 IMPORTANT SCIENTIFIC ISSUES THAT NEED TO BE ADDRESSED**

(Not necessarily in order of priority)

- a. Measurement of the radiation budget (up/down/net) in the stratosphere: high resolution wavelength dependence is desirable to investigate effects due to particle size, optical resonances, etc.
- b. Effect of change in temperature on formation and properties of PSCs: microphysics, heterogeneous chemistry, occurrences of metastable phases, etc. Modeling, in situ measurements, and laboratory studies are required.
- c. Modeling of the effect of PSCs on radiative transfer requiring measurements of PSC composition, refractive index, shape, phase, spatial variability. Both in situ measurement and laboratory studies must confirm physical properties.



- d. Modeling of the effect of change in stratospheric temperature on circulation, tropospheric exchange, residence time, etc.
- e. Effect of stratospheric aerosol composition, distribution and transport on properties of cirrus clouds (albedo, IR radiance, etc.) in the upper troposphere. Compare relative tropo/stratospheric influences. Nuclei from either source may initiate ice formation. Note: cirrus clouds are extremely important in the planetary radiation balance, particularly because of their great areal coverage. In this context it is important to determine the origins of cirrus cloud nuclei. We need to know the high-level versus low-level sources, in particular the stratospheric contribution and their role in controlling optical depth.
- f. Ozone depletion can affect the climate
  - (1) directly by re-distributing global temperatures.
  - (2) indirectly by increased ultraviolet (UV) radiation killing microorganisms in oceans, hence affecting dimethyl sulfide (DMS) production. As a consequence, the cloud condensation nuclei (CCN) concentration could be smaller and the cloud albedo could be lower.
- g. To mitigate the catastrophic effect of a major volcanic eruption, e.g., meteoric impact, a world-wide organization of measurement and monitoring stations (e.g., lidar network, etc.) needs to be in place; also modeling and predictive capabilities of climatic impact of large stratospheric disturbances, comparable to other emergency preparedness organizations (e.g., hurricanes and nuclear release), is required.
- h. We need to consider possible impact of space exploration activities, e.g., rocket exhaust emissions and debris from re-entering vehicles. This has to be compared with other external inputs such as:
  - (1) Micrometeors and cosmic dust
  - (2) Tropospheric injection
  - (3) Future increases in high-altitude aircraft operation
- i. There is evidence that aerosols produced in the troposphere or at the surface are transported into the stratosphere. Volcanic emissions are a clear example of this; deep convective storms may also be a mechanism, particularly in the tropics.

### 1.3 ROLE IN HETEROGENEOUS CHEMISTRY

Panel Members: S. Elliott (Leader), T. Deshler, P. Hamill, C. S. Kiang, S. Kinne, J. Rosen, L. Stefanutti, O. B. Toon, R. P. Turco, C. Wilson

Aerosol surfaces in the atmosphere permit chemical reactions to occur which would otherwise be kinetically unfavorable. In addition, they provide a mechanism for the removal of chemical species

from the gas phase through condensation and sedimentation. Reaction rates of heterogeneous processes may be dependent on the physical state and the chemical composition of particles. The alteration of aerosol composition by surface reactions can affect the refractive index and volume of the particle.

### 1.3.1 INTRODUCTION

Stratospheric particles can initiate heterogeneous reactions leading to major shifts in gas phase photochemistry. Catalysis of nitrate transformations on or in polar stratospheric clouds (PSCs) is now thought to be capable of inverting the standard relationships between chlorine reservoirs and  $\text{ClO}_x$  radicals in the polar stratospheres, and a direct consequence has been massive Antarctic ozone depletion. Sulfate particles could produce similar redistributions at lower latitudes. Dustsonde baselines indicate that the background aerosol may be increasing by as much as 5-10% per year, and with anthropogenic total chlorine concentrations rising simultaneously, studies of catalysis by sulfate would seem to be in order. It has also been demonstrated recently that injections of volcanic particulates may be connected to localized loss of ozone. Revival of plans for supersonic commercial aircraft promise to further enhance interest in aerosol reactions.

The following paragraphs give several areas in which the influence of stratospheric particles on chemistry is still being investigated, along with a set of suggestions for relevant experiments, including laboratory, in situ, and numerical approaches. The major thrust is toward the PSCs because they are of immediate and critical concern, but many of the overall concepts apply to sulfate as well. The problems presented are subdivided into sections focussing on the physical chemistry of heterogeneous reactions, aerosol properties bearing on them, and fundamental microphysical issues such as nucleation. Some broader scale objectives for future polar field campaigns are also discussed.

### 1.3.2 PHYSICAL CHEMISTRY OF HETEROGENEOUS TRANSFORMATIONS

The composition of polar stratospheric clouds is perhaps the most basic facet of their physical chemistry and can determine nitrate reactivity, but it remains poorly understood on several key levels. For example, the nitric acid trihydrate (NAT) which probably comprises the bulk of Type I PSC material can contain a wide range of water impurity concentrations. The water mole fraction in turn specifies hydrochloric acid (HCl) holding capacities and efficiencies for the reaction between nitrate and HCl. Actual water concentrations in the NAT particle, however, remain unknown. This workshop has yielded debate on the merits of in situ mass spectroscopy of the Type I PSC contents. An instrument capable of performing this spectroscopy is under development, and it may be possible to obtain rough measurements of solid phase  $\text{H}_2\text{O}/\text{HNO}_3$  ratios. One participant suggested that PSC crystals might be captured and analyzed wet chemically for the same information, but it was then argued that they were too unstable toward evaporation for this to be an effective strategy. In situ infrared spectroscopy of the Type I clouds could provide clues to their composition, but would have to be preceded by laboratory documentation of NAT absorption bands. Whether or not NAT can contain a wide range of water impurity concentration is somewhat controversial: NAT-ice mixtures undoubtedly exist, but there is no consensus as to whether these are mixtures with water dissolved in NAT (or vice versa) or if NAT and ice are in separate inclusions. Some laboratory data support the former but a Gibbs-Duhem analysis of NAT vapor pressures supports the latter.

Solid vapor pressures are closely linked with composition, but our understanding of them is again incomplete. The effects of impurities, coatings and simultaneous condensation of water and NAT have not been investigated. While partial pressures of  $\text{H}_2\text{O}$  and  $\text{HNO}_3$  are available for NAT alone, they have not been associated in detail with the water concentration of the lattice. Adsorptive thermodynamics is a theme related to PSC vapor pressures. An adsorption equilibrium constant defines the tendency for a molecule of interest, most particularly a heterogeneously active nitrate or acid, to cover the cloud particle surface. The equilibrium can in turn be converted into a surface binding enthalpy, which enters into calculation of rates for several elementary steps occurring early in the surface reaction process, including desorption and two dimensional diffusion. It could be profitable to undertake the determination of adsorption isotherms for the PSC heterogeneous reactants on NAT and ice. Preliminary values have already been reported for  $\text{HCl}$  which indicate the formation of two hydrogen bonds with the crystal. Nitric acid adsorption might also be accessible, but measurements for the nitrates would be complicated by competition from surface reactions.

Although a growing body of circumstantial evidence involving Lagrangian photochemical models points to Type I particles as the chief chlorine activators, several workshop participants felt that the PSC/ $\text{ClO}_x$  connection merits further study. In the laboratory, continued effort is needed in measuring collisional efficiencies for nitrate reactions on simulated PSC materials. Early experiments were plagued by improper characterization of solids. Studies of  $\text{ClONO}_2 + \text{HCl}$  on water ice, for example, were conducted at pressures orders of magnitude above the stability threshold for hydrochloric acid hydrates. NAT efficiencies may be more realistic at the moment, but the dependence of reactivity on  $\text{HCl}$  solubilities is not well established, and as mentioned above, stratospheric  $\text{HCl}$  mole fractions cannot be predicted with confidence as yet. Aerosol chlorine activation could perhaps be verified directly by monitoring air up- and downstream from a single cloud. The technical and political difficulties inherent in this proposition were considered at length, and there was debate on the wisdom of flying in or near mountain wave structures.

The possibility exists that certain subtleties of heterogeneous chemistry are being overlooked because attention has centered on the net chlorine activating reactions of  $\text{ClONO}_2$  and  $\text{N}_2\text{O}_5$ .  $\text{ClO}$  dimerization, for example, may be augmented on the PSC surfaces, and as the rate controlling step in an odd oxygen removal cycle, this recombination is clearly distinct from the nitrate transformations.  $\text{HO}_x$  species and their reservoirs are decoupled to some extent from the chlorine and nitrogen balances which limit ozone lifetimes, and so heterogeneous interactions of the hydrogen families remain unexplored. It would be valuable to study the thermodynamic and kinetic features required for significant heterogeneous catalysis of a gas phase reaction. The candidate processes highlighted by such a study could then be tested in a photochemical simulation. Another logical priority is detailed bridging of polar photochemical and microphysical calculations through a conceptual model of PSC surface processes.

### 1.3.3 PARTICLE PROPERTIES BEARING ON HETEROGENEOUS CHEMISTRY

Shape is an obvious physical property which contributes to the reactivity of nitrates or acids on stratospheric aerosols. An irregular surface exhibiting hopper systems or graininess will add to the available heterogeneous transformation area. There is evidence that at least some of the polar water ice particles are hexagonal and columnar, but departures from this familiar configuration are probable, and Type I clouds have not been intensely scrutinized. Replicator work provides qualitative



information on particle shapes. Polarization data recently permitted the discrimination of two new particle categories within the Type I designation, perhaps corresponding to diverging morphologies. Shape is of course a function of the physical state of the aerosols as well, and although polar air temperatures lie well below freezing temperatures for liquid water or aqueous nitric acid, there was some discussion on freezing point depression by unidentified impurities. Several workshop participants cautioned that it might be advisable to verify that the PSCs actually exist as solids, and several experimental ideas arose in response to this suggestion. Vapor samples near the condensation point could be cooled under controlled conditions and the particle formation process observed, either in the lab or perhaps even in situ. Particle radii are a related matter and many of the invited lectures centered on the reporting of size distributions. There was general agreement among the heterogeneous chemistry panel members that further monitoring of the sizes of stratospheric aerosols was essential, whether at mid-latitudes or the poles.

Particle optical effects are critical in calibration and operation of instruments used to study stratospheric aerosols, and consequently there was considerable interest in their clarification. The dependence of the solid refractive index on composition has not been delineated for PSC materials, and several participants felt that theoretical calculations would be desirable for the optics of non-spherical particles. Infrared spectroscopy of the Type I clouds could also be classified under this heading.

#### **1.3.4 RELEVANT MICROPHYSICS**

Major uncertainties permeate calculations of stratospheric aerosol production, especially with regard to energy barriers against nucleation. It is currently thought that background sulfate aerosols entering the polar stratosphere act as nuclei for Type I PSCs, and Type I in turn for Type II. Laboratory measurements of the saturation ratio necessary to achieve particle formation could improve the modelling situation for each step in this sequence. The initial background sulfate nuclei are present in the mid latitude stratosphere as liquid, and it has been presumed that the strong supercooling to which they are subjected in the polar vortices results in freezing. This is only conjecture, however, and laboratory verification could be enlightening.

#### **1.3.5 OTHER TOPICS**

The heterogeneous chemistry group outlined and had discussions on subjects such as denitrification and dehydration, chlorine activation, and the impact of different sorts of temperature variation on the microphysics. These areas were not considered in detail due to a lack of time.

#### **1.3.6. CONCLUSIONS AND SCIENTIFIC ISSUES**

- a. Mechanisms of the heterogeneous reactions are only beginning to draw attention, but are at the heart of several critical stratospheric problems. Chlorine activation is known to take place on particle surfaces at the poles, and potentially in other localities. Aspects of aerosol physical chemistry underlie the heterogeneous mechanisms. Compositions, vapor pressures, and in the PSC instance, adsorption equilibria, all pertain to aerosol reactivities and surface elementary steps.

- b. Fundamental physical properties including morphologies and size distributions also enter into the determination of heterogeneous reaction rates. Optical interactions are central to the measurement of these properties.
- c. The process of nucleation, through which the aerosols under consideration are formed originally, remains obscure at all stages for polar stratospheric clouds.



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## II. MEASUREMENTS

### 2.1 IN SITU MEASUREMENTS

Panel Members: Robert Stevens (Leader), Terry Deshler, Guy Ferry, Bernd Georgi, Jindra Goodman, Gunthard Metzger, Joe Pinto, Richard Santer, Ken Snetsinger, Chuck Wilson

#### 2.1.1 SUGGESTED MEASUREMENTS

To interpret the effect of aerosols and gases on the atmosphere, information on the vertical and horizontal distributions of aerosol particles, their size distributions and concentration of precursor gases is required.

The panel discussed trends in aerosol and gaseous components. It is important to know whether these are increasing or decreasing. The particulates which should be monitored for trends are sulfate particles, nitrate particles as well as graphitic carbon and organic non-graphitic carbon. Alumina particles should also be monitored. The minimal quantities of interest are total mass and number concentration. Gaseous species which should be monitored are  $\text{SO}_2$ ,  $\text{HNO}_3$ ,  $\text{HCl}$ ,  $\text{HBr}$ ,  $\text{COS}$ ,  $\text{HF}$ ,  $\text{CH}_3\text{SO}_4\text{H}$ , and  $\text{ClNO}_2$ .

Sources of stratospheric aerosol particles that need to be considered are emissions by aircraft, in particular supersonic aircraft operating in the stratosphere and the advection into the stratosphere of surface-generated man-made (urban and industrial operations) and natural (volcanic emissions, sea-salt, non sea-salt sulfate, desert dust) aerosols. The accumulation of debris from space operations is a distinct probability that deserves particular attention.

Optical properties of aerosols have to be assessed in relation to chemical composition, and physical state (phase, shape). Attention has to be given to a change of the physical and chemical properties as a function of particle size. This is important because the chemical composition determines the aerosols' refractive index.

The chemical properties of aerosols determine the freezing point and the reactivity properties of their surface.

The residence time of the aerosol has to be assessed in relation to chemical composition, particle size, meteorological processes in various seasons, and phoretic forces.

#### 2.1.2 MEASUREMENT TECHNIQUES

Measurements of aerosols and gases at frequent intervals and over vast areas of the atmosphere generally involve remote sensing techniques. These techniques depend on the optical properties of the species investigated. Optical properties can vary greatly with aerosol size and refractive index. Remote sensing instruments look at light scattering, extinction, depolarization, and reflection.

In situ sampling methods for aerosols and gases are also subject to difficulties in the interpretation of collected data. Instruments that sample aerosols and gases by bringing them into the sampling platform require that sampling is isokinetic and that surfaces are inert to prevent losses due to reactions with the inlet surface. Optical particle counters require knowledge of the refractive index and shape of the particle sampled. Most optical particle counters have a lower size limit of about 0.3 micron diameter with a few down to 0.1 micron diameter. The results are size distributions and total concentrations. Because the Mie scattering curve is not smooth, there are practical limits to the fineness of the size distributions. Chemical data is not measured by optical particle counters. CN counters can go to lower size, but provide only total number concentrations. Aerosols sampled by impaction on a surface or a filter permit an interaction between the surface and the particle; thus chemical or physical changes may occur prior to examination of the sampling surface. Volatile components of the aerosol are most easily lost as the temperature of the sample is often radically changed prior to analysis. Impaction instruments can provide chemical data but usually provide only limited time resolution and relatively few samples during a flight.

### **2.1.3 AIRBORNE MEASUREMENTS: RECENT ACCOMPLISHMENTS**

A number of recent NASA aircraft programs have made use of in situ aerosol measurement techniques to advance our understanding of stratospheric aerosols and important phenomena involving these aerosols. Several balloon experiments have been performed in Europe and in the United States; a brief summary of current balloon-borne measurement programs is listed in table 1.

#### **2.1.3.1 Aerosol Climatic Effects (ACE)**

In-situ measurements were made from the NASA Ames U-2 aircraft in the study of volcanic aerosols. The Ames Wire Impactor (AWI) and PMS laser particle spectrometers provided measurements of size distributions of the volcanically enhanced aerosols following El Chichon (1982). The condensation nucleus counter (CNC) data documented new particle formation in the stratosphere following the eruption of El Chichon and the Multiple Filter Sampler (MFS) documented sulfate loading.

#### **2.1.3.2 Stratosphere-Troposphere Exchange (STEP)**

Three aircraft were used in documenting tropopause folding in 1984 and in the tropics in 1987. The U-2 aircraft carried a CNC, the AWI, and the MFS. The MFS samples were analyzed for Be-7, a stratospheric tracer. Condensation nuclei (CN) provided a tracer for tropospheric air in the diagnosis of wave motion occurring above tropopause folds.

#### **2.1.3.3 Satellite Validation Experiments**

These experiments were flown to provide comparisons between remotely sensed satellite (SAM and SAGE) and in situ aerosol measurements. AWI and laser aerosol spectrometer size distributions from U-2 and ER-2 aircraft were compared with satellite occultation extinction measurements. Dustsondes were used in several locations; balloon-borne polarimetric and limb observations were performed in Europe in order to validate the SAGE II measurements.

**TABLE 1. CURRENT BALLOON-BORNE MEASUREMENT PROGRAMS OF STRATOSPHERIC QUANTITIES**

Group	Scientists	Measurements	Period	Frequency	Latitude
Univ. of Wyoming	Hoffmann, Rosen	Optical particle counter $r \geq 0.01, 0.15, 0.25 \mu\text{m}$	1972-1987	12/yr	41°N
Univ. of Wyoming	Hoffmann, Deshler	Optical particle counter $r \geq 0.01, 0.15, 0.25 \mu\text{m}$	1988-present	12/yr	41°N
Univ. of Lille (Aire sur Adour)	Herman, Santer	Photopolarimetry. Size distribution, refractive index, shape	1983-present	2/yr	46°N
Institute Spacial Belgique	Ackerman	Limb photographs from constant alt. balloon, at 3 colors, extinction profile asymmetry factor	1978-1985	1/yr	46°N
Max Planck Institute (Heidelberg)	Arnold	Ion mass spectrometer and cryo sampler	1984-present		46°N

**Polar Activities**

Univ. of Wyoming	Hoffmann, Rosen	Optical particle counter $r \geq 0.01, 0.15, 0.25 \mu\text{m}$	Dec-Jan 1974-1983	2-4/yr	78°S, 90°S
	Hoffmann, Rosen, Deshler	Optical particle counter, Ozone Large particle counter $r > .01$ to $5 \mu\text{m}$	Sept 1986- 1990	6/yr	78°S
	Rosen	Back scatter sonde Ozone	Jan 1989, 1990	10/yr	82°N
	Hoffmann, Arnold	Particle counter $r > 0.01-5 \mu\text{m}$ Ozone Mass spectrometer	Jan 1989, 1990, 88, 83	4/yr	
CHEOPS	Univ. of Lille	Photopolarimetry	1988, 1990	2/yr	68°N



#### **2.1.3.4 Airborne Antarctic Ozone Experiment (AAOE)**

Measurements were made in the ozone hole from the ER-2 aircraft. Measurements of aerosols with the AWI, the laser aerosol spectrometer, and the Reactive Odd Nitrogen Experiment confirmed the presence of nitrate on the aerosols in polar stratospheric clouds (PSCs). The first replication of ice crystals in the stratosphere was achieved with the Formvar Ice Crystal Replicator (ICR) in a polar stratospheric cloud. Sulfate particle formation in the polar vortex was observed with the CNC. Remote measurements of PSCs were made from the DC-8 aircraft with lidar. These measurements enhanced understanding of PSC structure, particle shape, and formation.

#### **2.1.3.5 Airborne Arctic Stratospheric Experiment (AASE)**

Aerosol instruments similar to those flown in AAOE were operated in AASE. These measurements added to our understanding of the physics and chemical composition of PSCs.

#### **2.1.3.6 Chemistry of the Ozone Polar Stratosphere (CHEOPS)**

Polar stratospheric clouds were studied by balloon-borne polarimetry during two European experiments in winter 1988 and 1990.

### **2.1.4 IN SITU MEASUREMENTS: RECOMMENDATIONS AND FUTURE RESEARCH CONSIDERATIONS**

Characterization and measurement of chemical and physical properties of stratospheric gases and aerosols requires intensive cooperation of the chemistry, physics, meteorology, and electronic and data processing fields which are discussed below. In addition, aerospace technology must be employed. Our recommendations are as follows:

#### **2.1.4.1 Chemistry**

The chemical composition, phase equilibria, surface properties and photochemical behavior of stratospheric aerosols are not clearly understood or adequately measured by current technologies. The following are areas where technology needs to be developed:

- (1) On-line elemental analysis (e.g., sulfur, nitrogen, halogens, and carbon).
- (2) Procedures to measure phase equilibrium states of species contributing to stratospheric chemical phenomena (e.g., nitric acid trihydrate from  $\text{HNO}_3$ ,  $\text{HCl}$  from aerosol chlorides).
- (3) Methods to determine the chemical and physical properties of aerosol and gaseous species in real time or in bulk samples.
- (4) Determination of the source of carbonaceous grains has been a major problem. Grains which contain minerals, depending on which phases are present, can be identified as extraterrestrial or terrestrial, anthropogenic or natural. However, carbonaceous grains originate from a variety of sources and no criteria have been developed for the unequivocal



identification of their various origins. It is known that carbonaceous materials from extraterrestrial sources commonly have isotopic anomalies in the hydrogen they contain. For example, an extraterrestrial carbon sample can have a per mil deuterium/hydrogen (D/H) enrichment (relative to terrestrial standards) of hundreds or even thousands. Fleming et al. and Blake et al. report the application of a new ion imaging technique to carbonaceous grains from the Johnson Space Center (JSC) curatorial collection. In this technique, a scanning ion beam is used to produce two dimensional images of D/H ratios which have a resolution exceeding  $2.0\ \mu\text{m}$  (particles smaller than this can be detected but their shape cannot be ascertained). It has been proposed to use this technique to measure the per mil enrichment of D in a statistically significant number of carbonaceous particles from the stratosphere. These data will provide a first estimate as to the ratio of extraterrestrial to terrestrial carbonaceous particles in the stratosphere. If the flux of extraterrestrial grains into the atmosphere remains constant over time, this ratio will provide a measure of the increase or decrease of terrestrial carbonaceous materials over time.

#### **2.1.4.2 Physics**

Instrumentation that measures the physical properties of aerosols that impact the chemical reactions, atmospheric radiation and conductivity in the stratosphere needs to be improved. For example, instrumentation is needed to address the following issues:

- (1) Determination of the aerodynamic, electrical, and optical properties of stratospheric aerosols;
- (2) Real time measurement of temperature and humidity profiles;
- (3) Design and development of an artifact-free aerosol and gas collection apparatus;
- (4) Construction of measurement systems (sensors) that obtain reliable, interpretable data independent of temperature and barometric pressure.

#### **2.1.4.3 Meteorology**

Forecasting and assessment of meteorological conditions which influence stratospheric conditions, ozone depletion conditions and/or climatic changes need to be improved for the following topics:

- (1) Reliable forecasting of meteorological conditions in the troposphere and stratosphere.
- (2) Evolution of polar transport models for the description of horizontal and vertical profiles.

#### **2.1.4.4 Electronics and Data Processing**

Development and coupling of sensor and data processing equipment that will distinguish signal from noise pulses in real time which may vary from instrument to instrument is needed. These developments must include data processing for the following types of equipment:

- (1) Temperature, pressure, and humidity.
- (2) Flight data (e.g., coordinates and flight speed).
- (3) Data from physical and chemical sensors (e.g., mass and optical spectrometers).

#### **2.1.4.5 Instruments and Techniques**

Instruments and techniques must be improved to expand on the number of chemical species that can be monitored to understand the chemical and physical stratospheric processes. New technologies to minimize artifact formation and to provide reliable measurements to support the stratospheric research are needed. In particular, the panel recommends:

- a. Develop artifact-free inlet system(s) which can be coupled to various platforms to improve the quality and accuracy of aerosol measurements.
- b. Design annular denuders for stratospheric applications, develop new denuder coatings to expand on the number of chemical species that may be measured, and develop annular denuder systems to examine the phase equilibria of key stratospheric chemicals. (Feasibility studies of denuder operations at the appropriate Mach, Reynolds and Sherwood numbers are necessary to prove their applicability to the stratosphere.)
- c. Develop and/or improve particle sizing instrument(s) to measure in real time and over integrated periods the chemical and physical properties of aerosols over a size range between 0.01 and 10  $\mu\text{m}$  in aerodynamic diameter.
- d. Expand the use of particle-size-fractioning impactors which can be coupled to denuders to obtain size-fractionated samples for subsequent analysis (e.g., elemental composition, ionic species composition, morphology and soot content).
- e. Develop methods to measure the nucleation rate via CNCs and advanced DMA technology.
- f. Improve the design of canister samples to obtain COS, SO<sub>2</sub>, CO, and CH<sub>4</sub> hydrocarbon and halocarbon analysis.

#### **2.1.4.6 Aerospace Technology**

Platforms to obtain stratospheric data need to be improved to accommodate new technology currently under development. In addition, these new platforms must permit monitoring additional chemical and physical properties of the troposphere and stratosphere. These developments must include:

- a. Aircraft designed as a flying laboratory to support both the monitoring equipment and the flight test of new systems. These platforms should be designed to accommodate investigators assigned to the operation, maintenance and testing of stratospheric monitoring and sensor hardware. These

aircraft should stay aloft for 6-12 hours at altitudes up to 12 km and with no geographic restrictions (e.g., polar flights).

- b. High altitude platforms which can be used for measurements above 12 km up to 40 km and temperatures to  $-100^{\circ}\text{C}$  need to be developed for characterization of chemical and physical properties of stratospheric gases and aerosols (ref. NASA CP-10041, 1990). These platforms may be balloons and various aircraft equipped with:

- (1) On line meteorological data acquisition equipment and associated telemetry hardware.
- (2) On line chemical and physical data acquisition and telemetry.
- (3) Space and design for artifact free sampling, collection and/or analysis of gases and aerosols.
- (4) Payload should be in the 500-1500 kg range.
- (5) Flight times must be between 6-60 hours.

#### 2.1.4.7 ER-2 Measurements to be Made

Within the Antarctic ozone hole (the South Polar Vortex), there are several halogen species of critical importance.

- a. Cl: Only column integrated remotely sensed measurements of HCl exist, taken by Farmer at McMurdo. There are no in situ measurements in the region of interest. HCl should be measured both within and outside the vortex. This will allow the partitioning of chlorine species to be converted to ClO (obtained by Anderson  $\sim 1.3$  ppb), HCl and HOCl. It's not clear which is larger, HCl or ClO (probably ClO), so it is important to improve understanding of the Cl chemistry. Information on particulate chloride is also needed in order to assess the role aerosols play in the ozone-halide chemistry.
- b. F: HF is a good tracer for air motions within the polar vortex. If air is sinking in the vortex in relation to outside, HF could show it because its concentration increases sharply with altitude (it is formed from the photolysis of the Freons). If air is sinking throughout the polar winter, it implies that at the beginning of Spring when ozone destruction occurs, the concentrations of reactive species are characteristics of higher altitudes. All that is available now are ground based spectroscopic measurements. It is important to show if F is taken up by the clouds to identify its origin unambiguously.
- c. N:  $\text{HNO}_3$ -NAT. The partitioning between gas and particle forms of these compounds has to be described quantitatively.

Although long integration times on the order of an hour are needed for the measurements due to the non-availability of a fast response instrument, useful budget studies could be done by integrated measurements over the flight path.



#### **2.1.4.8 Future Measurements and Improvements using Balloon Technology**

Balloon borne measurements provide a valuable complement to other airborne measurements of stratospheric aerosol. Compared to aircraft they are less expensive, are not severely limited by meteorological conditions (for small balloons), can reach altitudes not currently possible with aircraft, and there are no sampling difficulties associated with extracting samples from a high speed airstream. To provide temporal variation of dynamic quantities, measurements can be repeated every few days if necessary. Each balloon sounding provides a vertical profile over a single location with vertical resolution on the order of less than 100 m. Long duration sampling at a constant altitude is possible with ballast controlled balloons, and these can be used for repetitive sampling in a single air mass. Hot air balloon facilities exist which provide the possibility of constant altitude sampling for periods on the order of 50 days. A brief summary of current balloon borne measurement programs is listed in table 1.

#### **2.1.4.9 Future Measurements and Improvements in Instrumentation**

The major need appears to be for measurements in the polar regions because of the role of polar stratospheric clouds in the heterogeneous chemistry involved in ozone depletion. Required improvements in characterization of the stratospheric aerosol include determination of the refractive index and/or chemical composition, the shape of particles larger than 1 micrometer, and the concentration of particles larger than 10 micrometers. Measurement of the refractive index or chemical composition is important for calibration of particle counters as well as to provide information for predicting the layers and temperatures at which to expect polar stratospheric clouds. Particle shape information can be used to infer particle nucleation and growth history, and is also important to calculate the surface area distribution available for chemical reactions. For each of these measurements, several techniques can be used and are under development. Large particles can be measured by extending the range of particle counters and by nephelometry. Particle shape can be determined by particle capture and replication techniques, and by observing departures from Mie theory in the analysis of polarization and radiance measurements. Measurements of refractive index are possible using polarization or backscattering measurements. Efforts are also being made toward measuring the vertical profile of chemical species using an ion mass spectrometer. Coupling these measurements with aerosol size distribution measurements would again provide a useful characterization of polar stratospheric clouds.

In the mid-latitudes monitoring the stratospheric background aerosol needs to be continued as does the tracking of volcanic events. In the event of a major volcanic eruption, frequent sampling should be conducted with a view to determining the extent of ozone depletion by heterogeneous chemistry on volcanic aerosol. The possibility of flying a manifold of denuders in a payload to provide a coarse vertical profile of the chemical species needs to be investigated. Data for comparison with remote sensing techniques needs to be provided, and different data sets from balloon measurements should be intercompared. An experiment was described which will investigate the composition of aerosol particles. A pulsed laser will evaporate and ionize single particles, followed by a time-of-flight mass spectrometer. The particles will be coarsely sized by light scattering before the pulsed laser is fired. The instrument is being developed for in situ measurements on a high altitude aircraft.

We have to emphasize that a complete description of the stratospheric aerosol is impossible using any one measurement technique, and that consequently joint experiments are mandatory.

## **2.2 REMOTE SENSING**

Panel Members: H. Jäger (Leader), J. Livingston, P. Wang (for P. McCormick), R. Santer, L. Stefanutti, Jun Zhou.

### **2.2.1 INTRODUCTION**

The stratospheric aerosol remote sensing working group defined objectives, data products, and present and future roles of remote sensors in coordinated stratospheric aerosol measurement programs.

The basic terms "stratospheric aerosols" and "remote sensing" were discussed at length, which resulted in the following clarifications. Although the term "stratospheric aerosols" is typically used to refer to liquid or solid particles or some combination of the two phases, it is widely recognized that both the formation and the lifespan of these particles are closely tied to their microphysical interactions with the various gaseous species that make up the condensate. Hence, the group concluded that it is imperative that future stratospheric aerosol studies include measurements of both the particles and the gases in order to maximize the amount of useful information obtained. The term "remote sensing," as applied to stratospheric aerosols, refers to measurements of the optical properties of the interacting particles and gases by active (lidar) and passive (radiometric) techniques using ground-based, airborne, or spaceborne sensors. The primary advantage in using remote sensors to measure stratospheric aerosol properties is the extended capability to characterize the spatial and temporal distribution of the aerosols.

In general, measurements obtained by active and passive sensors are complementary. Existing ground-based and airborne lidar systems yield measurements with high temporal and range (altitude) resolution. Present satellite-borne sensors use passive techniques and, hence, cannot provide data that are as finely resolved in time or space as the lidar measurements, but they provide an unparalleled capability to monitor stratospheric aerosols on a global scale for long time periods. Future satellite-based active (lidar) and next-generation passive sensors will extend this capability.

### **2.2.2 OBJECTIVES**

Several key objectives were identified for future stratospheric aerosol studies that employ remote sensors. These are:

- a. To identify possible trends due to natural and anthropogenic sources.
- b. To monitor long-term spatial and temporal distributions of background stratospheric aerosols, and to document perturbations in particle size, number, and distribution due to volcanic eruptions.



- c. To characterize aerosol and gaseous transport and dispersion within the stratosphere, and to extend our knowledge of stratospheric-tropospheric exchange mechanisms. (This implies an improved understanding of atmospheric dynamics processes.)
- d. To quantify the interactions between particles and gases in order to understand better both the microphysics of gas-to-particle conversion and cloud (cirrus and polar stratospheric cloud [PSC]) formation and evolution.
- e. To determine the climatic implications of changes in the radiative properties of stratospheric aerosols.

## 2.2.3 DATA PRODUCTS

The working group identified several desirable general and specific data products. Some of these are already being measured, although not necessarily in the stratosphere or at desired stratospheric altitudes, by one or more of the ~130 lidar systems that are operational worldwide (see Second International Lidar Researchers Directory, NASA, August, 1989 and Fifteenth International Laser Radar Conference, Abstract of Papers Part I and II, Institute of Atmospheric Optics, Tomsk, USSR, 1990), or by various satellite-borne passive sensors.

From a global climate perspective, it is essential that remote sensors be able to measure such parameters as particle concentration, size distribution, phase (liquid or solid) and shape, meteorological variables such as density, temperature and wind, and concentrations and distributions of gases such as water vapor, ozone, nitrogen compounds, sulfur compounds, and chlorine compounds. In all likelihood, it will continue to be necessary to rely on in situ measurements for information on particle composition and mass. Climatically relevant optical data should include volume extinction and scattering coefficients, and effective particle size, volume, and surface area. Additionally, measurements of the angular scattering characteristics (i.e., the scattering phase function) of the particles are necessary for the correction of passive remote sensing measurements for radiative effects due to intervening atmospheric aerosols. Data are necessary also to quantify aerosol multiple scattering effects on transmitted solar radiation and on limb path radiometric measurements of both the transmitted solar radiation and the radiation emitted by atmospheric gases.

## 2.2.4 CAPABILITIES

Ground-based active (lidar) systems provide measurements of high temporal and altitude resolution. Ground-based passive systems still need to be further developed with respect to altitude resolution.

Satellite-borne instruments which use both active and passive techniques are suitable for providing long-term stratospheric aerosol measurements with near-global coverage. Solar occultation and limb scattering techniques are examples of passive instruments, whereas the lidar technique is an example of an active technique.

#### **2.2.4.1 Ground-Based Remote Sensing**

Aerosol lidars should provide height- and time-resolved measurements of atmospheric backscattering ratio (the ratio of the total aerosol plus molecular backscatter to the molecular backscatter component), depolarization ratio (the ratio of the backscattered return measured in a plane perpendicular to the emitted laser beam to that measured in a plane parallel to the emitted radiation), and some measure of multiple scattering. Such data are valuable both as aerosol monitoring devices and as atmospheric diagnostic tools. Where spherical particle shape and Mie theory can be invoked, the aerosol backscatter data can be combined with ancillary measurements of the aerosol size distribution and composition to yield inferred wavelength-dependent optical properties such as aerosol extinction, effective particle size, and layer mass, surface area, and volume. Differential absorption lidars (DIAL) can measure the concentration and distribution of gases such as ozone, nitrogen dioxide, sulfur dioxide and water vapor. Raman lidars are presently being used to measure tropospheric water vapor profiles, and have the potential to measure aerosol extinction profiles. Measurements of stratospheric and mesospheric molecular density and temperature are presently being made with single wavelength lidars (Rayleigh lidars) and multiwavelength lidars that also use Rayleigh backscattering. Wind can be measured by Doppler Mie and Doppler Rayleigh lidars. Coincident measurements of molecular and aerosol backscatter are provided by high spectral resolution lidar systems.

#### **2.2.4.2 Lidar In-Space Technology Experiment (LITE) - Active Remote Sensing**

Future satellite borne lidar programs on U.S. (NASA) and European (ESA) platforms like LASA (Lidar Atmospheric Sounder and Altimeter) or ATLID (Atmospheric Lidar) will be preceded by lidar experiments on manned spacecraft missions. Such experiments are ALEXIS (Atmospheric Lidar Experiment in Space) aboard the Spacelab or LITE. The airborne projects LEANDRE (France) and the LASE (Lidar Atmospheric Sensing Experiment) by NASA also aim at the future use of lidar on space platforms.

The Lidar In-Space Technology Experiment (LITE) is currently being developed by NASA for flight aboard the Shuttle in 1993. The measurement objectives of LITE are to measure profiles of atmospheric aerosols and molecular densities, and to determine the height of cloud tops and the planetary boundary layer. The instrument consists of a 1-joule laser transmitter, together with a 1-meter diameter reflecting telescope. The laser transmitter has a flashlamp pumped Neodymium-YAG laser operating at 10 Hz, with a beam divergence of 1.0 milliradian at wavelengths 1.064  $\mu\text{m}$ , 0.532  $\mu\text{m}$ , and 0.355  $\mu\text{m}$ . This one-week mission will be a proof of concept flight to show the capabilities of a spaceborne lidar. It is anticipated that a series of these Shuttle flights will follow with a long-term flight on an unmanned spacecraft near the year 2000.

#### **2.2.4.3 Satellite Solar Occultation Measurements - Passive Remote Sensing**

Since October 1978, NASA has launched a series of three satellite instruments for the measurement of aerosols; the Stratospheric Aerosol Measurement (SAM II) on Nimbus 7, and the Stratospheric Aerosol and Gas Experiments I and II (SAGE I and II) on the Application Explorer Mission 2 Satellite and Earth Radiation Budget Satellite, respectively. They all use the solar occultation technique to measure the sun's attenuated intensity through the earth's limb during each spacecraft sunrise and sunset. Table 2 summarizes these missions, their latitudinal coverage,

wavelengths, and the retrieved species for each. In addition, it also includes information on SAGE III which has been accepted for phase B studies for NASA's Earth Observing system. The simultaneously measured stratospheric aerosol extinction at seven wavelengths and, in addition, water vapor, ozone, nitrogen dioxide, chloride dioxide, and temperature by the SAGE III instrument will substantially enhance the information for study of the behavior of stratospheric aerosols, and the

**TABLE 2. SATELLITE LIMB EXTINCTION MEASUREMENTS**

Experiment	Satellite	Launch	Latitude Coverage	Wavelength ( $\mu\text{m}$ )	Primary Species
SAM II (Solar)	NIMBUS-7	Oct. 1978*	64°-80°N 64°-80°S	1.0	Aerosol
SAGE I (Solar)	AEM-2	Feb. 1979**	79°N-79°S	0.385 0.450 0.600 1.0	NO <sub>2</sub> Aerosol O <sub>3</sub> Aerosol
SAGE II (Solar)	ERBS	Oct. 1984*	80°N-80°S	0.385 0.448 0.453 0.525 0.600 0.940 1.0	Aerosol NO <sub>2</sub> Aerosol Aerosol O <sub>3</sub> H <sub>2</sub> O Aerosol
SAGE III (Proposed) (Solar)	Eos/NPOP Eos/SSF	1996	55°-70°N 60°-90°S 40°S-40°N	0.290 0.385 0.430-0.450 0.525 0.600 0.740-0.780 0.920-0.960 1.02 1.55	O <sub>3</sub> above 50 km Aerosol NO <sub>2</sub> , Aerosol Aerosol O <sub>3</sub> below 65 km O <sub>2</sub> /T, Aerosol H <sub>2</sub> O, Aerosol Aerosol Aerosol
(Lunar)		1998	90°S-90°N	0.380-0.420 0.430-0.450 0.640-0.680 0.740-0.780 0.920-0.960	OCIO NO <sub>2</sub> NO <sub>3</sub> O <sub>2</sub> /T H <sub>2</sub> O

\*Presently still operational.

\*\*Obtained data through November 1981.



effect of aerosol changes on stratospheric ozone and climate. SAGE III will also make measurements during lunar moonrise and moonsets allowing nighttime species (e.g., nitrogen trioxide) to be measured and, in addition, expand the coverage during each spacecraft revolution.

#### **2.2.4.4 Satellite Limb Scattering Technology – Passive Remote Sensing**

Through the use of limb scattering technology, the satellite instrument can provide the measurement coverage to a daily global one, which will be about a month for the solar occultation technique. The satellite instrument of the Solar Mesosphere Explorer (SME) is a typical example, which operated from 1982 to 1986. The difficulties of this technique are the complex retrieval scheme and the time consuming inversion analysis required, due to multiple scattering processes involved in radiance measurements. With the computers becoming larger and faster, this technique is desirable in the near future.

#### **2.2.4.5 Airborne Remote Sensing**

Active and passive remote sensing experiments are already flying on a number of research aircrafts. Airborne lidar systems are operated by NASA, in France by CNRS and CNES (LEANDRE project) and in Germany by DLR (ALEX = Aerosol Lidar Experiment).

A solar spectrometer (sun photometer) is presently being studied for installation on a NASA ER-2 aircraft, which reaches altitudes of 20 km, for measurement of stratospheric optical depth contributions from aerosols, ozone and nitrogen dioxide. This instrument is being designed to obtain correlative measurements for validation of the SAGE III sensor, which will be located on NASA's Earth Observing System. The ER-2 spectrometer will consist of a grating/array detector arrangement and will take measurements at wavelengths between 350 nm and 1550 nm.

### **2.2.5 VALIDATION OF SATELLITE REMOTE SENSING**

The validation of satellite remote sensing requires coordinated correlative measurements under stable atmospheric conditions and spatial and temporal coincidence. The difficulty with aerosol validation is that different instruments generally measure different aerosol parameters. Therefore, satellite validation processes may involve data conversion and require appropriate aerosol models. In this regard, it is desirable to incorporate in the correlative measurements a set of aerosol instruments capable of providing detailed information, sometimes redundant, including as much information as possible on size distribution, refractive index, composition, and shape. This information is not only important for testing the aerosol model used for data conversion, but also crucial to the understanding of stratospheric aerosols. Table 3 summarizes the correlative sensors used in the worldwide SAM II, SAGE I and II measurement validation programs. The SAM II and SAGE I and II series of satellite experiments have shown themselves capable of providing long-term observations. Periodic correlative observations are required, therefore, for assuring that data quality/validation is unchanged, or for understanding any changes observed.



**TABLE 3. SENSORS USED IN SAM II, SAGE I AND II COMPARATIVE STUDIES**

Satellite Instrument	Correlative Sensor	Particle Parameter	Platform/Site
SAM II <sup>a</sup>	Dustsonde (2 Channels) Lidar Quartz Crystal Microbalance (QCM)	Number Backscatter (0.694 $\mu\text{m}$ ) Number, Mass, Composition	Balloon P-3 Sabreliner
SAGE I <sup>b,c</sup>	Dustsonde (2 Channels) Lidar Multifilter Wire Impactor (75 $\mu\text{m}$ ) QCM Integrating Plate Nephelometer	Number Backscatter (0.694 $\mu\text{m}$ ) Sulfate/Mass Number/Mass Number, Mass, Composition Absorption ( $\sim 0.55 \mu\text{m}$ ) Scatter Phase Function	Balloon P-3 U-2 U-2 Sabreliner, U-2 U-2 Sabreliner
SAGE II <sup>d</sup>	Dustsonde (3/6 Channels) Lidar Wire Impactor (75 $\mu\text{m}$ , 0.5 mm) Particle Measurement System laser probes (PMS) Multifilter QCM Lidar <sup>e</sup>  Limb Photography <sup>e</sup>  Polarimetry <sup>e</sup>	Number Backscatter (0.523, 0.69 $\mu\text{m}$ ) Number/Mass  Number/Mass  Sulfate/Mass Number, Mass, Composition Backscatter (0.694 $\mu\text{m}$ ) Backscatter (0.532 $\mu\text{m}$ ) Backscatter (0.532 $\mu\text{m}$ ) Backscatter (0.532 $\mu\text{m}$ ) Scatter Radiance (0.84, 0.44, 0.375 $\mu\text{m}$ )/Ext. Polarization Diagram (0.85, 1.65 $\mu\text{m}$ )/Size Parameters	Balloon P-3 U-2  U-2  U-2 U-2 Garmisch, FRG OHP, France Frascati, Italy Florence, Italy Balloon/Belgium  Balloon/France

<sup>a</sup>Russell et al., 1981, p. 41.

<sup>b</sup>McCormick et al., 1981, p. 41.

<sup>c</sup>Russell et al., 1984, p. 41.

<sup>d</sup>Russell and McCormick, 1989, p. 41.

<sup>e</sup>Ackerman et al., 1979, p. 41.

## 2.2.6 FUTURE REQUIREMENTS

Satellite solar occultation measurements have the unique capability to provide global and permanent surveys of stratospheric aerosols and have played a leading role in tracing volcanic effluents and in the discovery of polar stratospheric clouds. They must be pursued by the launch of the

Stratospheric Aerosol and Gas Experiment (SAGE) III sensor as are Earth Observing System (EOS) sensor as soon as possible to avoid a discontinuity of the excellent record of SAGE II observations.

Since the early observations in 1964, lidars have played a major role in the detection of background stratospheric aerosols and the monitoring of stratospheric aerosol distribution changes due to volcanic eruptions such as El Chichon. During the past decade, lidars have been shown to be equally valuable as correlative sensors in major stratospheric aerosol satellite validation programs such as Stratospheric Aerosol Measurement (SAM) II, Stratospheric Aerosol and Gas Experiment (SAGE) I and II, and in extensive polar stratospheric measurement field campaigns such as the National Ozone Expedition (NOZE) I, II, III, and NASA's Airborne Antarctic Ozone Experiment (AAOE) and Airborne Arctic Stratospheric Expedition (AASE). It is the consensus of the remote sensing working group that lidar will continue to be an invaluable tool for stratospheric aerosol measurement and satellite validation. Recognizing this, the group identified a number of key needs that need to be addressed by the worldwide lidar community to ensure the maximum utilization of lidar measurement capabilities in future international stratospheric measurement programs:

- a. Additional lidars with the minimum capability to sense background stratospheric aerosols need to be established in the high northern latitudes with temporary access to the arctic winter polar vortex, the southern mid-latitudes, and the southern hemisphere within the austral winter polar vortex area. This goal is consistent with the goals of a global network for the detection of stratospheric change, as recently formulated;
- b. For lidars participating in such a network, a set of minimum lidar instrumental requirements, such as wavelength, transmit energy, telescope receiver size, eye safety standards, etc., might be defined;
- c. Lidar measurement analysis procedures and data products (including archival formats and access procedures) need to be standardized. This might include definition both of standard aerosol or gas quantities (such as aerosol scattering ratio or gaseous concentration) to be reported and also a set of standard altitudes at which these data are to be reported. Also included would be strict guidelines for performing lidar signal normalization, extinction (transmittance) corrections to measured backscatter signals, instrumental error corrections, and uncertainty analyses;
- d. There is also a need for further development of remote sensing tools like advanced lidar techniques such as Doppler and high resolution lidar and microwave remote sensing.

### III. RECOMMENDATIONS

Stratospheric aerosols play a role in global climate change through their effects on the Earth's radiation balance and by affecting upper tropospheric and polar stratospheric cloud radiative and chemical properties. They also provide the nuclei on which type I and II polar stratospheric cloud particles can form. In addition, they are part of the heterogeneous chemical system by which ozone is being destroyed in the polar and possibly the mid-latitudinal stratospheres. Because of the importance of stratospheric aerosols, we make the following recommendations.

- a. Continue the programs of satellite solar occultation observations, with possibly more aerosol channels to improve the size distribution retrieval.
- b. Set up a lidar network of some 20 stations over regions as large as the United States to study tropospheric exchanges of water vapor, and gaseous and particulate sulfur compounds. A dedicated theoretical program of stratospheric dynamics should go along in parallel with the observational program.
- c. Use high-altitude long-lived (one month or longer) balloons, equipped with solar-powered transmitters tracked by satellites, to supplement the lidar program.
- d. Measure the radiation budget (up-down-net) with high resolution spectral radiometers in unison with in situ determination of physical and chemical aerosol characteristics.
- e. Measure the effects of changes in temperature on the formation and properties (microphysics, heterogeneous chemistry, occurrence of metastable phases) of polar stratospheric clouds in situ and in the laboratory.
- f. Investigate the effects of composition and transport of stratospheric aerosols on the physical/chemical (composition, phase, shape, size) and optical (albedo, IR radiance) properties of upper tropospheric cirrus clouds.
- g. Assess the possible climatic impacts of space exploration activities, e.g., rocket exhaust emissions, debris from re-entering spacecraft in the context of the effects of other external inputs, e.g., micrometeors and cosmic dust and tropospheric injections due to, e.g., high-altitude aircraft operations.
- h. Establish long-term stratospheric aerosol records for studies of long-term variations over point locations.
- i. Combine lidar and satellite measurements to form a more complete monitoring system. Alone, neither has all of the desirable features, but together they form a compatible system for the following reasons:



- (1) There are gaps in the satellite data because of satellite orbital characteristics.
  - (2) Failed satellites will produce time gaps in the data; the lidar network is an inexpensive backup.
  - (3) The lidar backscatter properties of aerosols along with the satellite extinction properties of the aerosol contain more information on the aerosol optical properties than can be provided by a single type of measurement. Spectral lidar measurements would be better yet.
  - (4) Fresh injections of volcanic aerosols can produce such a large horizontal extinction along the SAGE horizontal path of view that the measurement method will not work well, or even not at all, because of multiple scattering or complete extinction. Lidar, on the other hand, can still be used.
  - (5) To quantify the effects of aerosols on remote sensing of surface and atmospheric properties, more experiments are needed. Lidar will provide the best data for aerosol profile directly above experiment sites.
- j. Satellite validation work by ground- and aircraft-based sensors should continue. If the stratospheric aerosol is changing, data from all measurement methods, in situ and remote, will be needed to verify conclusively the characteristics of the change(s) seen.
- k. In situ measurements are a vital link to combine with both satellite and lidar measurements. Neither satellites nor lidar can provide information on the composition of the lowest part of the stratosphere. Satellites do not have the vertical resolution and lidars cannot yet measure much in the way of chemical composition. Therefore it is mandatory to
- (1) Develop artifact-free inlet systems for aircraft and balloons which can be coupled to measurement systems to improve the quality and accuracy of aerosol measurements.
  - (2) Develop and/or improve particle sizing instruments (particle-size-fractionating impactors) which can be coupled to instruments (denuders) which identify chemical species.
  - (3) Develop and apply technologies (time of flight mass spectrometer) to do on-line elemental analysis (sulfur, nitrogen, halogen, carbon), to measure phase equilibrium studies of species contributing to stratospheric chemical phenomena (nitric acid trihydrate from  $\text{HNO}_3$ ,  $\text{HCl}$  from aerosol chlorides), and to determine the physical properties of aerosol and gaseous species in real time.
- l. Platforms to obtain stratospheric data need to be improved to accommodate new technologies currently under development. These must include
- (1) Aircraft designed as a flying laboratory to support both the monitoring equipment and the flight test of new systems. These platforms should be designed to accommodate investigators assigned to the operation, maintenance and testing of stratospheric monitoring and



sensor hardware. These aircraft should stay aloft for 6-12 hours at altitudes up to 12 km and with no geographic restrictions (e.g., polar flights).

- (2) High altitude platforms which can be used for measurements above 12 km up to 40 km and temperatures to  $-100^{\circ}\text{C}$  need to be developed for characterization of chemical and physical properties of stratospheric gases and aerosols.
- m. Airborne sunphotometry is recommended to measure multi-wavelength optical depths, which are a fundamental input to aerosol/climate interaction models.

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## APPENDIX

## ABSTRACTS

**ANNULAR DENUDERS FOR USE IN GLOBAL CLIMATE  
AND STRATOSPHERIC MEASUREMENTS  
OF ACIDIC GASES AND PARTICLES**

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Measurements of acidic (e.g.,  $\text{SO}_2$ ,  $\text{HNO}_3$ ) and basic gases (e.g.,  $\text{NH}_3$ ) that coexist with fine particles ( $<2.5 \mu\text{m}$ ) may be useful for determining the impact of these species on global climate changes and determining species (e.g.,  $\text{HNO}_3$  and  $\text{HNO}_3$  trihydrate) that influence stratospheric ozone levels. Annular denuders are well suited for this purpose. A new concentric annular denuder system, consisting of a three-channel denuder, a Teflon-coated cyclone preseparator, and a multistage filter pack has been developed, evaluated, and shown to provide reliable atmospheric measurements of  $\text{SO}_2$ ,  $\text{HNO}_2$ ,  $\text{HNO}_3$ ,  $\text{NH}_3$ ,  $\text{SO}_4^{2-}$ ,  $\text{NH}_4^+$ ,  $\text{NO}_3^-$  and  $\text{H}^+$ . For example the precision of the annular denuder for the ambient measurements of  $\text{HNO}_3$  and nitrates at concentrations between 0.1 to  $3 \mu\text{g}/\text{m}^3$  was  $\pm 12$  and 16%, respectively. The 120 mm  $\times$  25 mm three-channel denuder is encased in a stainless steel sheath and has annular spaces that are 1 mm wide. This design has been shown to have nearly identical capacity for removal of  $\text{SO}_2$  as conventional 210 mm  $\times$  25 mm single-channel denuder configurations. The cyclone preseparator was designed and tested to have a  $D_{50}$  cutoff diameter of  $2.5 \mu\text{m}$  and minimal retention of  $\text{HNO}_3$ . The annular denuder system could be designed and configured for measurements of stratospheric concentrations of  $\text{HNO}_3$  and the trihydrate of nitric acid. The system would be arranged to measure simultaneously  $\text{HNO}_3 + \text{HNO}_3 \cdot 3\text{H}_2\text{O}$  and  $\text{HNO}_3$ . The difference would be  $\text{HNO}_3 \cdot 3\text{H}_2\text{O}$ . The  $\text{HNO}_3$  would be removed with a  $\text{Na}_2\text{CO}_3$  treated denuder. The  $\text{HNO}_3$  and trihydrate would be collected on  $\text{Na}_2\text{CO}_3$  treated filter. Tests simulating stratospheric conditions would be needed to optimize the annular denuder/filter pack coatings and configuration prior to field testing the system in a UR-2 type aircraft.

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**ANNULAR DENUDEUR SYSTEMS:  
PRECISION AND APPLICABILITY TO STRATOSPHERIC SAMPLING**

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Annular denuder systems (ADSs, e.g., fig. A1), are designed to sample both gases and particles from ambient air without cross interference. Researchers at the University of Kansas have been using ADSs to detect chemical gradients over rural forest canopies and have developed accurate and precise methods for the sampling and analysis of low concentrations of gaseous and particulate species over short (2-4 hr) sampling periods. Figure A2 shows typical analytical precision data for nitrate and sulfate. Table A1 presents both sampling and analytical precision data for a 2-hr test using 4 collocated ADSs. On this particular day, the overall precision for  $\text{HNO}_3$  was unusually poor. The overall precision for the major constituents of interest is generally about 3-5% for typical ambient concentrations.

ADSs could potentially be used to determine the concentrations of  $\text{HNO}_3$  and fine-particle nitrate in the stratosphere, and to simultaneously characterize the chemical composition of stratospheric aerosols. Assuming 50 mb pressure, a 2-hr sampling time, a 50 Lpm sampling rate, and a low blank value, an overall precision of about 15% should be possible for 1 ppbv of  $\text{HNO}_3$  or nitrate. One possible system (designed to avoid the use of external power) is illustrated in figure A3. Systems employing heating could be used to sample volatile fine-particle nitrate, which would include nitric-acid trihydrate (NAT); and pump-driven systems could accommodate more complex systems involving denuders and filter packs in sequence. Alternate coatings could possibly be developed to selectively sample species such as  $\text{Cl}_2$  and  $\text{ClONO}_2$ . ADSs might also be useful in laboratory studies of heterogeneous chemical reactions involving NAT aerosols.

**TABLE A1. TWO-HOUR FIELD TEST RESULTS USING FOUR COLLOCATED ADS SAMPLERS<sup>a</sup>**

Constituent	Aqueous Concentration, $\mu\text{g/L}$					Amb. Conc.
	Sample 1	Sample 2	Sample 3	Sample 4	Average	$\mu\text{g/m}^3$
Coarse-particle nitrate-N						
Bent-tube inlet	$31.6 \pm 0.9$	$37.4 \pm 1.7$	$33.8 \pm 0.8$	$33.8 \pm 0.5$	$34.2 \pm 2.4$	$0.228 \pm 7.0\%$
Impaction disk	$60.8 \pm 0.5$	$62.6 \pm 0.8$	$63.2 \pm 0.9$	$61.2 \pm 0.6$	$61.9 \pm 1.1$	$0.413 \pm 1.8\%$
Total	92.4	100.0	97.0	95.0	$96.1 \pm 3.2$	$0.640 \pm 3.3\%$
Coarse-particle sulfate						
Bent-tube inlet	$41.0 \pm 3.1$	$41.8 \pm 3.4$	$26.3 \pm 1.2$	$34.8 \pm 2.6$	$36.0 \pm 19.9$	$0.240 \pm 55.3\%$
Impaction disk	$15.9 \pm 3.1$	$19.9 \pm 2.4$	$18.2 \pm 2.4$	$20.6 \pm 5.6$	$18.6 \pm 2.4$	$0.124 \pm 12.9\%$
Total	57.9	61.7	44.5	55.4	$54.9 \pm 7.4$	$0.364 \pm 13.5\%$
$\text{HNO}_3\text{-N}$	$51.3 \pm 0.0$	$42.3 \pm 1.6$	$52.7 \pm 1.1$	$43.9 \pm 1.1$	$47.5 \pm 5.2$	$0.317 \pm 10.9\%$
$\text{HNO}_2\text{-N}$	$0.0 \pm 0.0$	$0.0 \pm 0.0$	$0.0 \pm 0.0$	$0.0 \pm 0.0$	$0.0 \pm 0.0$	$0.000 \pm 0.0\%$
$\text{SO}_2$ (as sulfate)	$1575 \pm 19$	$1511 \pm 28$	$1564 \pm 40$	$1559 \pm 7$	$1552 \pm 28$	$10.347 \pm 1.8\%$
Fine-particle nitrate-N	$73.1 \pm 1.3$	$74.9 \pm 1.7$	$80.3 \pm 0.7$	$78.3 \pm 3.3$	$76.7 \pm 3.3$	$0.511 \pm 4.3\%$
Fine-particle sulfate	$956 \pm 20$	$972 \pm 12$	$1012 \pm 15$	$983 \pm 3$	$981 \pm 24$	$6.54 \pm 2.4\%$

<sup>a</sup>Ambient air sampled for 2.0 hours at 12.5 Lpm; 10-mL extracts; standard deviations shown in  $\mu\text{g/L}$  for individual samples (5 replicates by ion chromatography) and 4-sample averages, and as a percentage for average ambient concentrations.



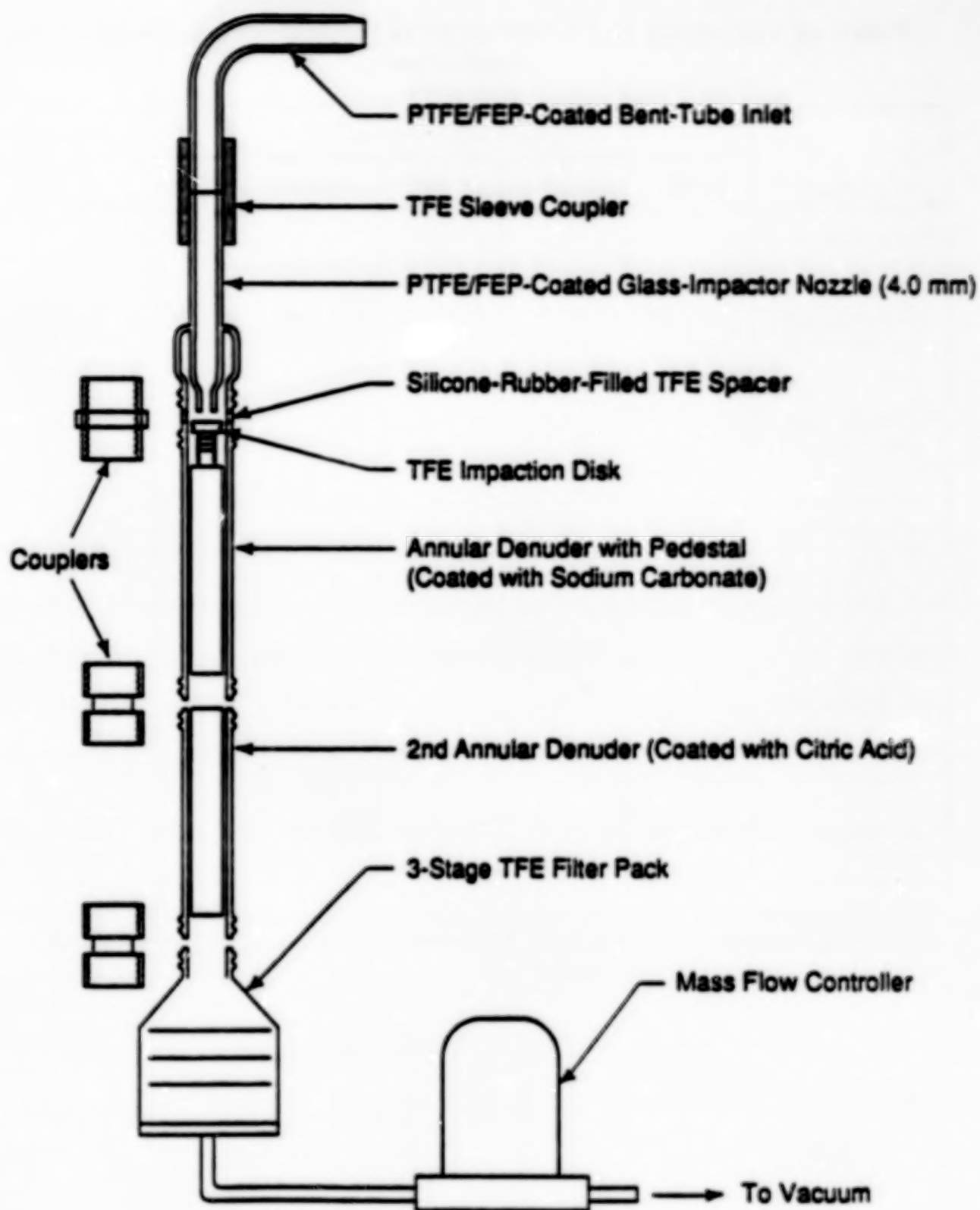


Figure A1. Annular denuder system.

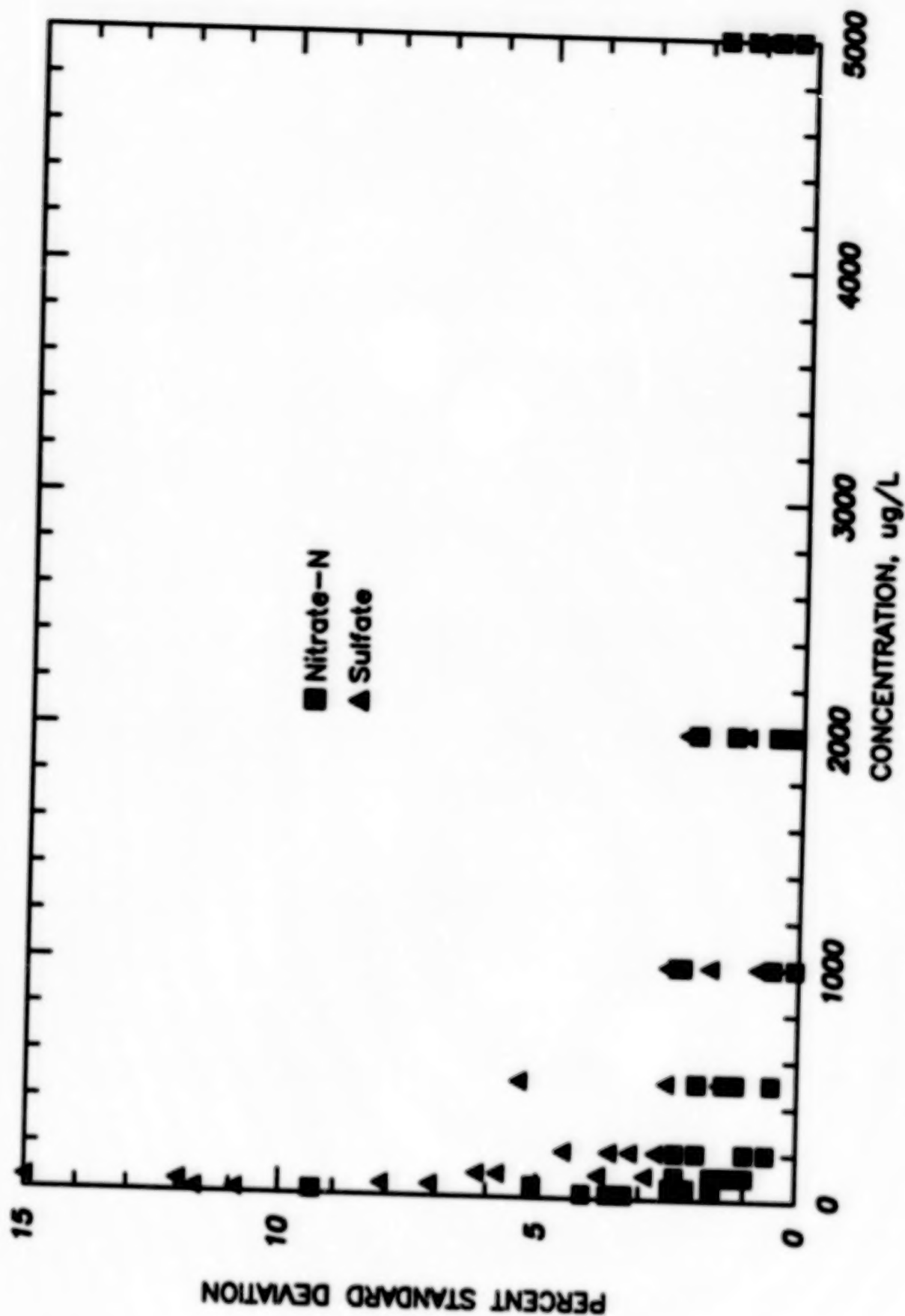


Figure A2. Precision of ion chromatographic analysis of nitrite and sulfate (each data point is based on 8 to 10 injections).

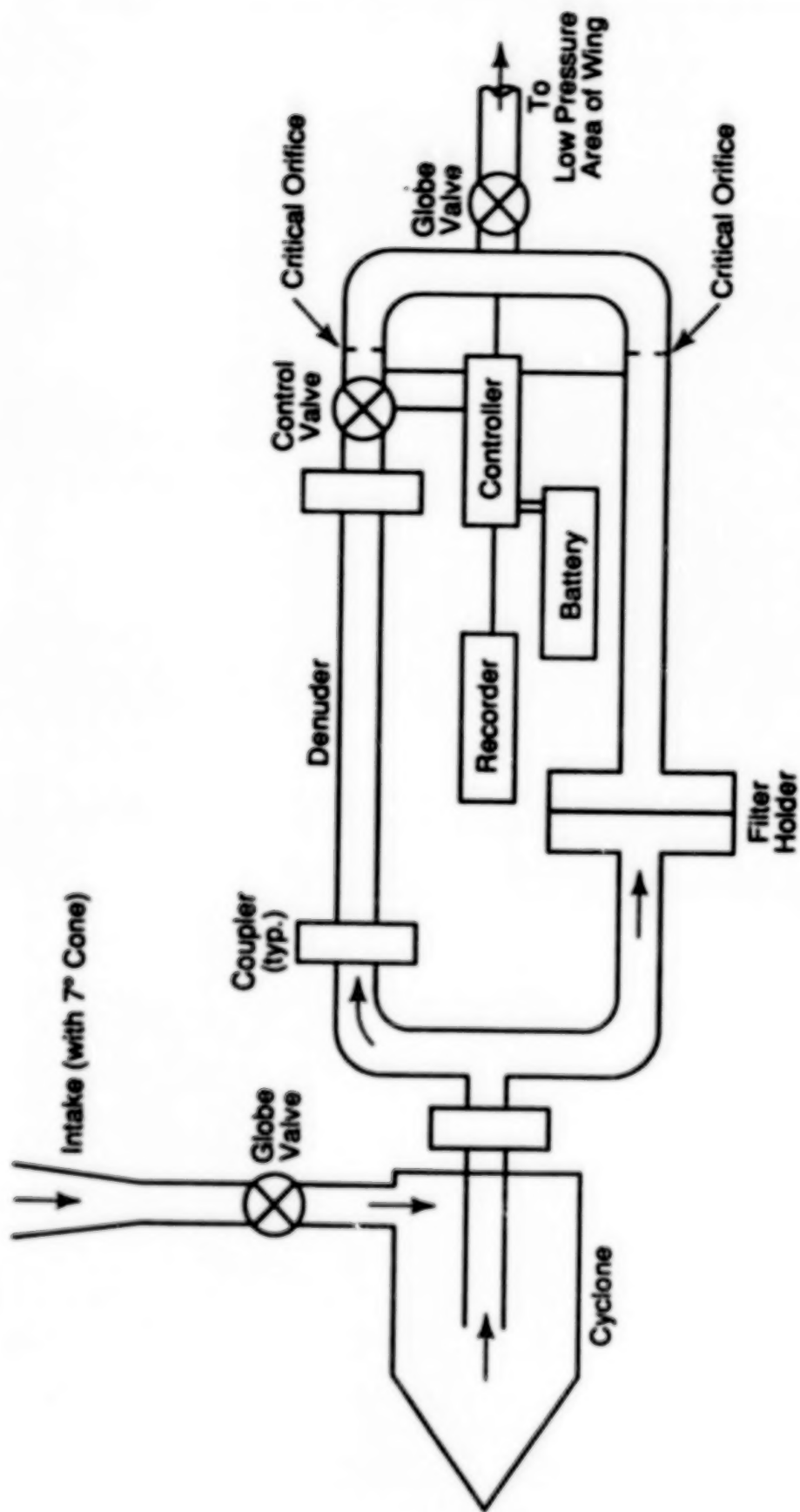


Figure A3. A Ram-powered annular denuder system (ADS) potentially useful for sampling stratospheric aerosols.

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**LABORATORY AND FIELD STUDIES OF STRATOSPHERIC AEROSOLS: PHASE  
CHANGES UNDER HIGH SUPERSATURATION**

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It is well known that water in the form of isolated small droplets supercool as much as 40°C below their equilibrium melting point. Solutions similarly supercool (with respect to water) and supersaturate (with respect of the solute). Experiments are described in which bulk solutions typical of atmospheric aerosols (nitric acid, sulfuric acid, and hydrates; ammonium sulfate; ammonium bisulfate; sodium chloride) are supercooled and/or supersaturated and nucleated to initiate crystal growth. Supersaturation of 300% is readily attainable, with linear growth of crystals increasing roughly as (supercooling/supersaturation)<sup>2</sup>. The implication of the experiments is that the situation of metastability in polar stratosphere clouds is very likely, with nucleation only occurring under a high degree of supercooling or supersaturation.

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**MEASUREMENTS IN POLAR STRATOSPHERIC CLOUDS OVER ANTARCTICA IN  
SEPTEMBER 1989**

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The results of 6 balloon flights at McMurdo Station, Antarctica, under varying temperature conditions, are used in a study of polar stratospheric clouds during September 1989. A new particle counter, with size resolution in the  $0.5\text{ }\mu\text{m}$  radius region, indicates that cloud size distributions are always bimodal. Mode radii ranging from  $0.05$  to  $0.10\text{ }\mu\text{m}$  were observed for the small particle mode, representing the sulfate layer or condensational growth enhancements of it. The data are not inconsistent with the expected increase in size with decreasing temperature of the small particle mode in the sulfate layer owing to deliquescence although this phenomenon is often masked by nitric acid trihydrate (NAT) condensation when temperatures are sufficiently low. Mode radii generally ranged from  $1.5$  to  $3.5\text{ }\mu\text{m}$  for the large particle mode at concentrations 3 to 4 orders of magnitude lower than the small particle mode. The large particle mode, which normally comprises most of the mass, is presumably caused by NAT condensation on larger particles of the sulfate layer and indicates  $\text{HNO}_3$  mixing ratios of 1 to 5 ppbv for most of the cloud layers observed, suggesting substantial denitrification. On several occasions, distributions were observed with mode radii as high as  $7\text{ }\mu\text{m}$ , and correspondingly large inferred mass, indicating water ice clouds in the 12 to 15 km region. On other occasions, absence of such clouds at very low temperatures indicated water vapor mixing ratios of less than 3 ppmv suggesting dehydration. Generally the inferred  $\text{HNO}_3$  mixing ratios were higher in the lower stratosphere, suggesting redistribution through particle sedimentation.

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**VALIDATION OF LIDAR AND SATELLITE REMOTE SENSING**

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Validation of observations is an important part of any remote sensing program. With respect to aerosols this is exceedingly difficult because each technique typically measures a different aerosol parameter. Correlative sensors, therefore, must be capable of measuring the same parameter over the same size range or include enough measurements to properly characterize the aerosol, possibly with the assistance of models. In any correlative measurement program, simultaneity in space and time is required. This can be less restrictive during periods of atmospheric stability. In this presentation, the discussion will be focused on the validation programs developed for NASA's Stratospheric Aerosol Measurement (SAM II), and the Stratospheric Aerosol and Gas Experiments I and II (SAGE I and II). To date these have been limited to the stratosphere. Since satellite sensors are capable of providing near-global and long-term observations, periodic correlative observations over various global locations and seasons are required for assuring that data quality is unchanged, or for understanding any changes observed.

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## LIDAR MEASUREMENTS AT GARMISCH, CONVERSION MODEL AND MIDLATITUDE AEROSOL EFFECTS

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Lidar backscatter soundings of the stratospheric aerosol layer have been routinely made at the IFU, Garmisch-Partenkirchen (47.5°N, 11.1°E), since October 1976. The period until 1979 was characterized by a very low level of stratospheric particulate load. After this background period significant perturbations of the stratosphere were observed after the eruptions of Mount St. Helens (1980), Alaid (1981), Nyamuragira (1981), El Chichón (1982), and Nevado del Ruiz (1985). A volcanically quiescent period followed until the Redoubt eruption at the end of 1989.

Determinations of particle extinction, mass and surface area of the stratospheric particulate load are important in climatic considerations and heterogeneous chemistry effects. Balloon-borne particle counter measurements of the University of Wyoming are used to derive height and time resolved conversion factors to convert lidar backscatter data to extinction, mass and surface area values. This is done by combining particle size distributions based on the Wyoming data with Mie backscatter and extinction functions. These conversion factors are applicable to midlatitude lidar observations and have been used to convert ruby lidar measurements at the IFU.

Following the El Chichón eruption abnormal stratospheric ozone deficiencies were observed above northern extratropical stations during winter 1982/83. Ozone variations measured by ozonesondes at Hohenpeissenberg (37 km from Garmisch) were analyzed and compared to particle surface values derived from the IFU lidar measurements. It was found that surface area densities above  $5\text{--}10\ \mu\text{m}^2\text{cm}^{-3}$  correlate with ozone minima which are not transport related. The analysis reveals that up to 40% of the observed ozone deficiency, depending on altitude, appears to be related to the perturbation caused by the El Chichón eruption.

A comparison of the volcanically quiescent periods 1977/78 and 1987/89 shows that the non-volcanic stratospheric background load had increased. This observation indicates the possibility that anthropogenic sulfur emissions to the stratosphere might be increasing which would have implications for the radiation balance and for stratospheric chemistry.

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## **POLAR STRATOSPHERIC CLOUD MEASUREMENTS BY MEANS OF DEPOLARIZATION LIDAR IN THE ANTARCTIC**

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POLE (Polar Ozone Lidar Experiment) is a cooperative project between the French Service d'Aeronomie of CNRS (S. Godin, G. Megie) and the Italian IROE-CNR (L. Stefanutti, M. Morandi, M. Del Guasta). The project is supported by the Italian Program for Antarctic Research (PNRA) and the French Territoires Antarctiques et Australes Francaises (TAAF). This program has started with the implementation of a first depolarization backscattering lidar [1] at the French Antarctic base of Dumont d'Urville (66°S, 140°E) in January 1989, for measurements both of background stratospheric aerosols and Polar Stratospheric clouds. Measurements have been carried out on a routine basis for 2 years, with almost one measurement per day in the period of absence of PSCs and with long term measurements in the occurrence of PSCs [2,3].

This program comprises also the installation in January 1991 of a complex Ozone lidar which will replace the backscattering system and extend its measurements to tropospheric and stratospheric Ozone and to stratospheric and mesospheric temperatures. This new system, operating with 5 wavelengths and 6 receiving channels allows also the measurement of the backscattered and depolarized signal produced by PSCs and background aerosols [4].

The location of Dumont d'Urville is peripheral in respect of the Polar Vortex and from the 2 years measurements it appears that only for reduced periods the station lies below it. From the stratospheric lidar measurements carried out in these 2 years, from a first analysis it results that no indication of background aerosol loading may be evidenced during such a short period of time, this also due to the uncertainty in the calibration of the lidar under low aerosol loading [2].

Polar Stratospheric clouds have appeared over Dumont d'Urville in the months of July and August; sporadic events have been noticed in September 1989, while no evidence of PSCs was in the same month of 1990.

The depolarization technique seemed to be quite efficient in the detection of different types of PSCs. Evidence of PSCs of Type Ia and Ib was recorded in several occasions in 1989, with the detection of clouds presenting high (above 2) scattering ratios and very low depolarization ratios (practically no depolarization) and cases which presented lower scattering ratios but much larger depolarization ratios (larger than 10%). One example is shown in Fig. A4. Not all PSCs measurements could be easily classified as in the case above. In order to obtain a better classification of PSCs types good temperature information are necessary. Not always such information were available as in 1989 in many occasions the balloons carrying the Rowinsondes would explode at altitudes of 16-17 km, thus making it impossible to have some good information of layers located up to 23 km. A better and more reliable technique for launching the balloons has been implemented since September 1989. Long sequel of stratospheric profiles carried out at a few minutes interval, for several hours, permit to evidence the evolution of PSCs over the lidar station.

Finally a long term program, with routine measurements carried out over several years seems the best solution for a better understanding of the polar stratosphere.

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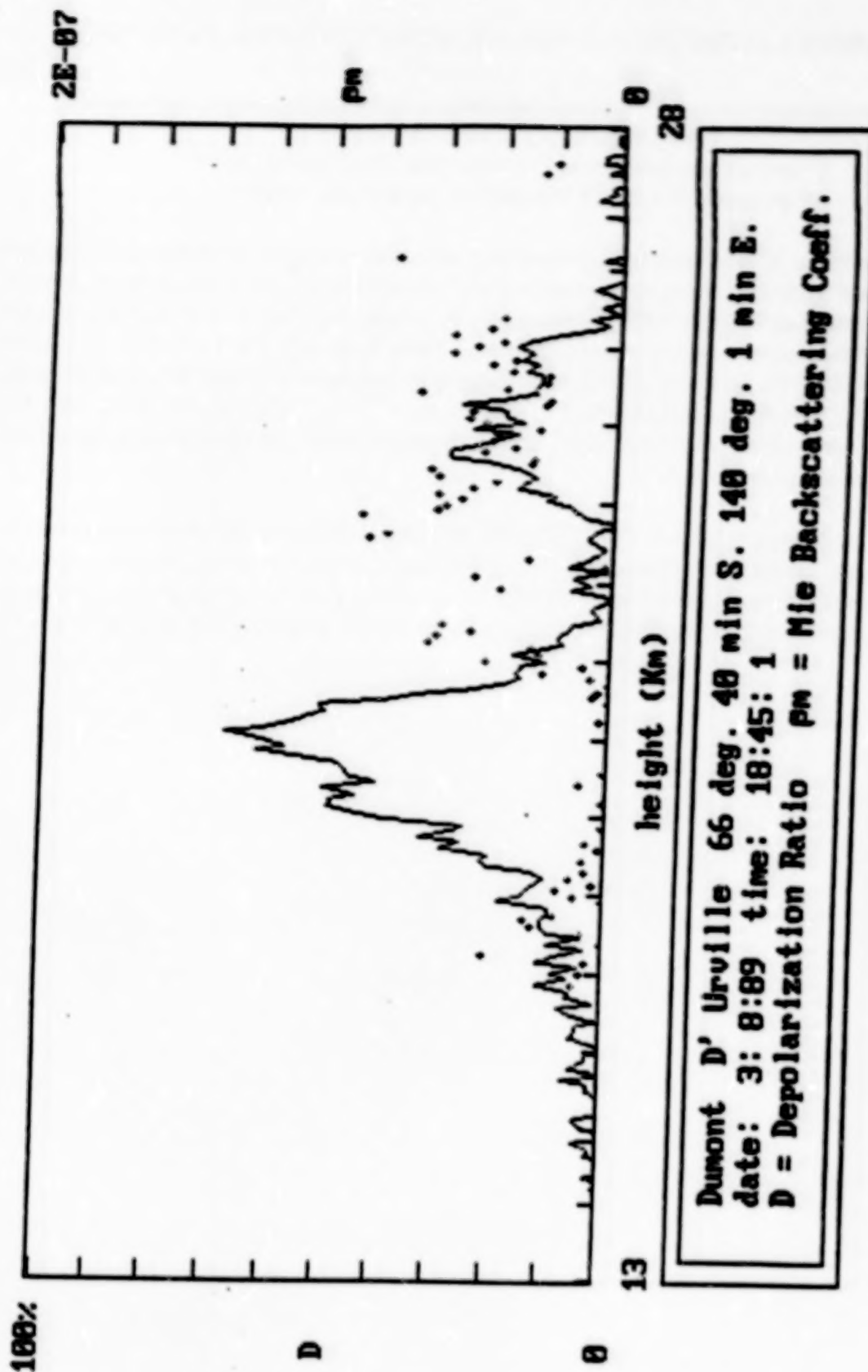


Fig. A4. Depolarization ratio (Mie crossed polarized component versus the sum of the parallel and the perpendicular Mie component)-dots, and the Mie backscattering coefficient-continuous line versus height.

## COMPARISON BETWEEN BALLOON POLARIMETRY AND SAM II/SAGE II

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The balloon borne infrared polarimeter described in R. Santer's paper provides optical tangent depths at 850 and 1650 nm. Using the aerosol model derived from the polarization measurements, they are interpolated at 1000 or 1020 nm for comparison with the SAM II and SAGE II tangent depths at 1000/1020 nm obtained by solar occultation. Three balloon flights from Aire sur l'Adour ( $44^{\circ}\text{N} - 0^{\circ}$ ) on 04-22-85, 10-12-85 and 04-21-86, have been compared with SAGE II profiles within 600 km from the launch site and one flight from Kiruna ( $68^{\circ}\text{N} - 20^{\circ}\text{E}$ ) on 01-28-88 with two SAM II profiles on both sides of the balloon. The agreement is within the error bars, in the altitude range (12-25 km) of the balloon data.

From the four aerosol channels of SAGE II (385, 450, 525 and 1020 nm) it is possible to retrieve information on the aerosol size distribution, i.e. an effective radius and an effective radiance, which have been compared with the values retrieved from the balloon polarization diagrams; the agreement is found good up to 22 km; above disagreements appear which can be due to the poorer quality of the data.



## EFFECTS OF STRATOSPHERIC AEROSOLS ON CLIMATE. OVERVIEW

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The primary effect of stratospheric aerosols is a modification of the radiation budget, mostly by scattering (and possibly absorption) of the solar radiation; the aerosols also influence slightly the longwave radiation. The resulting effect on climate is made complex by various feedbacks with chemistry and dynamics.

A simple modelling of the planetary albedo change due to an increase of the stratospheric aerosol content allows to clarify the influence of the various aerosols parameters (optical depth, single scattering albedo and asymmetry factors), and of the ground albedo. In most cases, the aerosol layer increases the albedo leading to a cooling of the lower atmosphere and of the ground, with a warming in the stratosphere itself.

After a large volcanic eruption, as Agung or el Chichon the aerosol optical depth increases from the background value of about 0.003 to values larger than 0.1. The impact of a volcanic eruption on temperature has been modelled and compared to observations after Agung eruption (Hansen et al., 1978) and better documented after El Chichon; temperatures at 30 mb have been found a few degrees higher in 1982 than the previous year average (Labitzke et al., 1983).

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## AEROSOL SENSING FROM BALLOONS USING PHOTOPOLARIMETRY

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A photopolarimeter, designed to measure the radiance and the polarization ratio at  $\lambda = 0,85 \mu\text{m}$  and  $\lambda = 1,65 \mu\text{m}$ , is set up on a gondola of a stratospheric balloon. By rotating the gondola, measurements of the diffuse solar light are obtained for a whole range of the scattering angles. An inversion scheme, based on the assumption of a log-normal size distribution, provides the relevant size parameters, the refractive index and the slant optical thicknesses of the aerosols.

Since 1983, this experiment was launched twice a year from Aire Sur Adour (S-E of France). The post El-Chichon stratospheric aerosol proved to be quite stable in size and nature (hydratid sulfuric acid), but of the abundance decreased of by a factor 10 between 1983 and 1987.

The experiment was also launched to observe the stratosphere during the Arctic winter. Two flights, on January 28, 1988 and January 18, 1990, corresponded to an unperturbed stratosphere. But during a third flight, on 04/02/90, the temperatures were low enough for PCS formations. A multilayered situation was observed with (i) the standard  $\text{H}_2\text{SO}_4$  aerosols below 20 km, (ii) a dense layer, peaked at around 23 km, with scattering coefficients about 50 times layer than in background conditions, and (iii) and an upper layer rather sharp ( $\sim 1$  km), with micron sized particles.

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## THE EFFECTS OF SULFATE AND SOOT IN SMOKE ON ATMOSPHERIC EXTINCTION

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There was a big forest fire in Northeastern China in May 1987. An apparently new aerosol layer was detected at the base of the stratosphere on data of lidar (at Mauna Loa, Hawaii) and balloon (at Laramie, Wyoming). Large forest fire that burned from about 6-28 May 1987 produced extensive clouds of smoke that were tracked for thousands of Km on NOAA weather satellite imagery. It was found during and just after this fire concentration of particles in upper tropopause and lower stratosphere above Pacific ocean and Northern America increased in several times, even ten times. This increasing may be caused by the smoke of this forest fire. Integrated NRBS coefficients aloft Mauna Loa in stratosphere on 19 and 26 May 1987 were 1.4 and 1.3 times respectively of those on 7 May 1987. It was found and known that the increasing of atmospheric extinction coefficient was contributed mostly by sulfate and soot of particles from biomass burning and coal combustion. It was shown that their contributions can reach 80-90% of the whole extinction coefficient. Soot is the product of incomplete burning. The black carbon in soot feature a high light absorption property. Sulfate is mainly produced by  $\text{SO}_2$  transformation and feature a high light scattering ability. It was understood that the forest fire which inject smoke to high altitude of the atmosphere can increase atmospheric extinction and may lead to a climate perturbation.

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## **A LIDAR EQUIPPED WITH PHOTON COUNTER FOR MEASURING STRATOSPHERIC AEROSOLS**

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A large lidar system has been developed at the Anhui Institute of Optics and Fine Mechanics (31.31°N, 117.17°E). It is mainly composed of a high-power frequency-doubled Nd:YAG laser (120 mJ at 532 nm, 1 mrad), a 651-mm-diameter receiving telescope, and a photon counter (64 channel). The lidar observations are controlled by an IBM PC/AT computer. One of our projects is to use the lidar system to make regular observations of stratospheric aerosols and the profiles of scattering ratio have been obtained since November 1989. Meanwhile, a Xe-Cl laser (308 nm wavelength) is assembled to allow the lidar to take measurements of vertical ozone distributions.

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## IN SITU MEASUREMENTS OF LIGHT EXTINCTION OF STRATOSPHERIC AEROSOLS

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The extinction coefficient of ambient aerosol particles is measured using a multiple traverse cell (White Cell) with an effective path length of 100m. Measurements are performed at seven fixed wavelengths in the visible region using a white light source and an interference filter set with 2 nm bandwidth and centerwavelengths of 405.5, 450, 500, 550, 600, 650, and 692.5 nm. The total air flow in the system is 16.7 l/min, the volume of the chamber is close to 10 liter. It takes about one minute to fill the chamber with particles homogeneously, but it needs up to five minutes to get the chamber particle free.

Before measuring the aerosol, during two minutes the transmission of particle free ambient air is determined; then the aerosol passes through the chamber for a period of ten minutes and after this the transmission of particle free ambient air is measured again for eight minutes. All times are subject to change. At present the measurements are done with a frequency of 1 Hz but an increase up to 30 Hz is possible. The lower detection limit of the used White Cell is  $3.4 \times 10^{-6}/\text{m}$ . This is sufficient for measuring the extinction coefficient during most tropospheric and some stratospheric conditions. But of course, it will be necessary to increase the sensitivity a factor of ten when measurements under the clearest stratospheric conditions are planned.



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16. Abstract <p>Following a mandate by the International Aerosol Climatology Program operating under the auspices of International Association of Meteorology and Atmospheric Physics International Radiation Commission, 45 scientists from five nations convened in March 1990 at NASA Ames Research Center to discuss relevant issues associated with the measurements, properties and effects of stratospheric aerosols. This report is a summary of the discussions on formation and evolution, transport and fate, effects on climate, role in heterogeneous chemistry, and validation of lidar and satellite remote sensing of stratospheric aerosols. Measurements are recommended of the natural (background) and the volcanically enhanced aerosol (sulfuric acid and silica particles), the exhaust of shuttle, civil aviation and supersonic aircraft operations (alumina, soot and ice particles), and polar stratospheric clouds (ice, condensed nitric and hydrochloric acids).</p>					
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